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(71) Applicant *(for all designated States except US)*: **AQUA-MARIJN HOLDING BV** [NL/NL]; Beatrixlaan 2, NL-7255 DB Hengelo (NL).

(72) Inventors; and
(75) Inventors/Applicants *(for US only)*: **RIJN VAN, Cornelis Johannes Maria** [NL/NL]; Beatrixlaan 2, NL-7255 DB Hengelo (NL). **NIJDAM, Wietze** [NL/NL]; Berkelkade 11, NL-7201 JE Zutphen (NL).

(74) Agent: **JILDERDA, Anne, Ayolt**; Octrooibureau LIOC, P.O. Box 13363, NL-3507 LJ Utrecht (NL).

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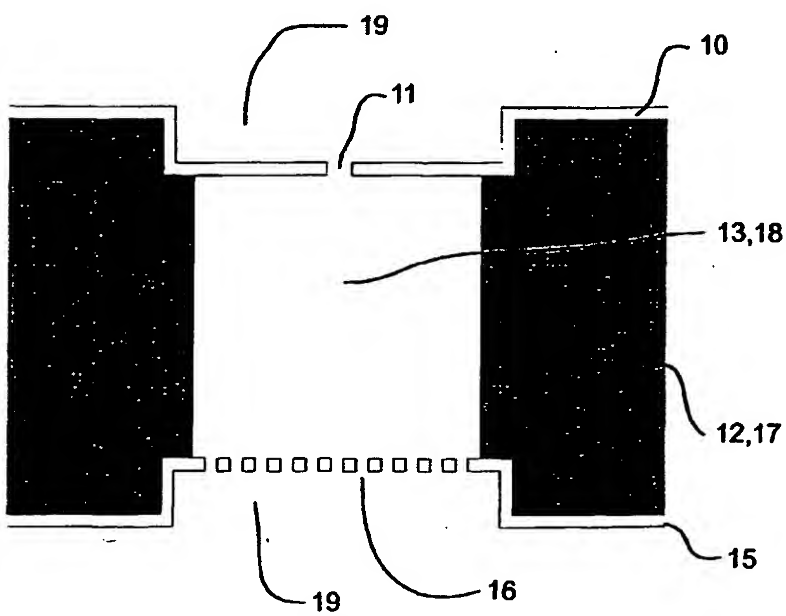
(54) Title: NOZZLE DEVICE AND NOZZLE FOR ATOMISATION AND/OR FILTRATION AND METHODS FOR USING THE SAME



(57) Abstract: Nozzle device and nozzle for atomisation and/or filtration as well as methods for using the same. The present invention relates to a nozzle and nozzle device for atomisation, in particular a micro-machined reinforced nozzle plate, that may produce small liquid droplets in air (spray) or into a liquid (emulsion) with a narrow droplet size distribution and to make small air bubbles into a liquid (foam) and to methods of making the same. The invention is further related to a nozzle part for filtration as well as means and methods to facilitate atomisation and filtration.

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NOZZLE DEVICE AND NOZZLE FOR ATOMISATION AND/OR FILTRATION AND METHODS FOR USING THE SAME

The present invention relates to a nozzle device having a nozzle for atomisation of a fluid, the nozzle comprising a nozzle plate support body having a cavity extending from a first main surface to a second main surface thereof, and comprising a nozzle plate having at least one nozzle orifice in fluid communication with said cavity at said first main surface side of said nozzle plate support body. The invention further relates to a nozzles as used in such a nozzle device.

These devices are used for filtration purposes and or for atomisation of a fluid to produce small liquid droplets in air (spray) or into a liquid (emulsion) with a relatively narrow droplet size distribution and to make small air bubbles into a liquid (foam) and to methods of using the same. The device and especially the nozzle plate may be produced by micro-machining (Micro System Technology) which means that the subject nozzle part means are produced using lithography steps related to semiconductor fabrication methods.Alternatively spark erosion and laser drilling technigues may be used, but in general these tend to be less reproducible and less precise in comparison with micro-machining methods.

The performance of many atomisation devices can be improved if the atomising device provides very small droplets with a very narrow pore size distribution. For example, small droplets between 2 and 3 micron in diameter improve the effectiveness of medical atomisers because of the high (80%) deposition intake deep into the lungs. Also the stability of an emulsion (o/w, w/o) is greatly improved if the emulsion droplets are all of equal size. Besides that, the structural and rheological properties of many foams in the dairy industry can be improved by the use of very small air bubbles with a narrow size distribution.

The disadvantage of many conventional atomising devices is that they break bulk liquid or gas into relatively large droplets through use of stirring or turbulence. By more input of energy the large droplets will be broken up in smaller droplets. As the droplets become smaller than 20-

100 microns, they become harder to break and secondary atomisation typically ceases. The droplet size distribution is in most cases rather broad.

It is known from fuel injectors that nozzle structures may be used for obtaining a very fine spray for combustion improvement. Such small nozzle structures however are very sensitive for fouling and unwanted leakage due to blocked nozzle orifices. For a high throughput of equally sized droplets normally an array of identical nozzles is used. However if one or more nozzle orifices becomes blocked the size distribution will broaden. If a nozzle orifice becomes smaller through partial blockage the droplets of this orifice will also become smaller. Moreover if the blockage is very severe spraying(or jetting) will cease and liquid will flow through this orifice over the surface of the nozzle structure hence influencing or inhibiting spraying behaviour of the other orifices.

It is also known that very small nozzles suffer from a threshold pressure (Pascal pressure/capillary forces) before they start spraying. The threshold pressure is inversely proportional to the nozzle diameter. For a nozzle with a diameter of 1 micron this pressure is typical 1-3 bar. For an array of nozzles it is therefore very important that all nozzles have an equal geometry with narrow tolerances and that the threshold pressure is kept as low as possible.

A high flow rate can be achieved by choosing the flow resistance of each nozzle orifice as small as possible and/or by increasing the pressure difference over the orifice during jetting. Practically the jetting pressures are chosen to be fairly higher than typical 5-10 bar. Such pressures will exert high forces on the nozzle plate. The nozzle plate is therefore chosen fairly thick (> 4-5 micron) in order to withstand such forces. However a thick nozzle plate implies a long orifice length and thus a high flow resistance and subsequently a reduced flow rate.

SUMMARY OF THE INVENTION

It is inter alia an object of the invention to provide a nozzle device and a nozzle of the type referred to in the opeing paragraph in which these drawbacks have been counteracted at least to an impressive exntend.

To this end a nozzle device as described in the opening paragraph is according to the invention characterized in that said support body is provided with filtration means which comprise a filtration plate which is in fluid communication with said cavity at said second main surface side of said nozzle plate support body.

A further object of the present invention is to produce a properly constructed nozzle plate for atomisation at operational pressures smaller than 10 bar.

Another object of the present invention is to provide nozzle plates that produce droplets typically with a mean diameter of 10 micron or smaller with a very narrow droplet distribution.

Yet another object of the present invention is to provide nozzle plates for small handheld atomising devices with a throughput nearly independent of the viscosity of the fluid (e.g. medicine) and means to reproducible facilitate atomisation.

Yet another object of the present invention is to produce a properly constructed nozzle plate (filtration membrane) for filtration of small and large amounts of liquid or gas and means to facilitate filtration with such a filtration membrane, which may be used in combination with atomisation applications.

Yet another object of the invention is to provide nozzle plates for large atomising devices capable of substantial throughput of atomised liquid or gas.

Yet another object of the invention is to provide nozzle plates with orifices with a reduced flow resistance that can withstand high operational pressures.

Yet another object of the invention is to provide atomising devices that are rather insensitive for microbiological fouling and unwanted leakage due to blocked nozzle orifices.

Yet another object of the invention is to provide atomising devices that are less sensitive for the Pascal threshold pressure.

These and additional objects and advantages of the invention will become apparent from the technical description which follows.

It is to be understood that both the foregoing summary and the following technical description are exemplary and explanatory and are not restrictive of the invention as claimed.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig.1	is a cross section of a nozzle device with a nozzle plate and pre-filter for atomisation.
Fig.2	is a cross section of a nozzle device with a nozzle plate and pre-filter for atomisation, in which the nozzle plate and the pre filter are deepened for protection.
Fig.3	is a cross section of a nozzle device with a nozzle plate and pre-filter for atomisation made from one piece.
Fig.4	is a top view of a nozzle plate containing slits plus special slits for pressure reduction.
Fig.5A	is a cross section of a nozzle device containing more orifices to increase the throughput.
Fig.5B	is a cross section of a nozzle device containing more orifices to increase the throughput with the possibility of liquid flow on both sides of the membrane.
Fig.6	is a top view of closely packed nozzle plates.
Fig.7A	is a top view of interconnected nozzle plates.
Fig.7B	is a cross section of interconnected nozzle plates.
Fig.8A	is a cross section of a thick nozzle plate with reduced flow resistance.

Fig.8B	is a cross section of a spiral nozzle orifice.
Fig.8C	is a cross section of the manufacturing method of a spiral nozzle orifice.
Fig.9	is a top view of nozzle plate with improved jetting behaviour.
Fig.10A	is a top view of a substantially stronger nozzle plate with slit type orifices.
Fig.10B	is a top view of a substantially stronger nozzle plate with circular orifices.
Fig.11	is a cross section of a nozzle device with a coated nozzle plate.
Fig.12	is a top view of a nozzle plate with separated hydrophilic/hydrophobic membrane coating for improved jetting/jet start.
Fig.13	is a cross section of a nozzle plate with slightly protruding nozzles.
Fig.14	is a cross section of a nozzle plate with hydrophilic/hydrophobic coatings around the orifices.
Fig.15	is a cross section of a vibrating nozzle plate with drain plate.
Fig.16	is a cross section of nozzle plate, placed under an angle, in a cross flow channel.
Fig.17	is a top view of a nozzle plate with extra nozzles for co-flow.
Fig.18	is a cross section of a nozzle plate for emulsification.
Fig.19	is a cross section high performance filter with the possibility for light to pass through.
Fig.20	is a top view of a filter for analysing particles that isolates the particles for enhanced recognition.
Fig.21	is a top view of a plastic disc supporting a nozzle plate for analysis purposes.
Fig.22	is a cross section of a plastic disc supporting a nozzle plate for analysis purposes plus funnel.
Fig.23	is a cross section of a reusable nozzle plate with a transparent cross flow channel.
Fig.24	is a top view of a glass module plate for high volume nozzle plates.
Fig.25	is a cross section of a glass module plate for high volume nozzle plates showing the shallow cross flow channels.
Fig.26	is a cross section of a glass module plate for high volume nozzle plates showing a channel of the comb structure.

Fig.27A is a cross section of stacked module plates.

Fig.27B is a opened 3D view of stacked module plates showing the liquid flow within the stack.

Fig.28 is cross section of a nozzle plate for evaporation purposes.

TECHNICAL DESCRIPTION OF THE PREFERRRED EMBODIMENTS

A first embodiment of a nozzle device **1** is shown in **FIG. 1**. The nozzle device **1** comprises a nozzle for atomisation **2**, with a nozzle plate **10** with at least one nozzle orifice **11** and a nozzle plate support body **12** with a nozzle cavity **13**, further comprising filtration means **3** with a filtration plate **15** with at least one filtration orifice **16** and a filtration plate support body **17** with at least one filtration cavity **18**. This nozzle device **1** is rather insensitive for microbiological fouling and unwanted leakage due to blocked nozzle orifices **11** because of the placement of a pre-filter for the nozzle for atomisation **2**. Basically the nozzle for atomisation **1** and filtration means **2** are made with the same micro machining techniques giving many additional advantages. The two parts **2** and **3** may have similar size and flatness and can therefor easily be directly bonded or glued **22** together without the need of separate or elaborated connection parts, that may introduce particle contamination between the nozzle plate **11** for atomisation and filtration plate **15**. A silicon wafer containing a number of nozzles for atomisation and a silicon wafer with a number of filtration means may be first bonded together before sawing the wafer sandwich into separate dies with individual nozzle and filtration means.

Another embodiment of a nozzle device **1** is shown in **FIG. 2**, characterised in that the nozzle orifices **11, 16** are made in a 2-200 micron deepened region **19** of the nozzle plate **2,3** with respect to the nozzle plate support body **12** and filtration plate support body **17**, herewith protecting the nozzle and/or filtration means during manufacturing and assembly against scratches etc. With preference the nozzle plate support body **12** and filtration plate support body **17** are identical, **FIG. 3**, the cavities **13,18** are then made by etching the support material directly through the nozzle and filtration orifices **11,16**. With this manufacturing

process there can not be any particle contamination between the filtration plate 15 and the nozzle plate for atomisation 10. In some cases it has been proven useful to make in the filtration means 3 one or more filtration orifices substantially larger (1-3 micron) than the other ones (0.2-0.8 micron) in order to reduce the Pascal pressure or to facilitate the removal of etching material(gas). Alternatively the filtration orifices 16 are slit-shaped to reduce the Pascal pressure. A special embodiment of such a filtration plate has a number of very long slit shaped filtration orificess 20 (e.g. with a length of 50-100 micron) near the edges of the filtration plate. Depending on the width 21 of the filtration plate 15 (e.g. 100 micron) and the applied pressure (e.g. 1 bar) this slit will open due to local bending of the filtration plate 15 (FIG. 4). Preferably the fluid resistance of the filtration plate 15 is minimal 3 times smaller than the fluid resistance of the nozzle plate 10. By this the pressure across the nozzle device 1 is effectively only used for atomisation.

Nozzles for atomisation 2 can be made with known micro machining techniques. A mono crystalline silicon wafer 12 with thickness 400 micron is provided with a Low Pressure Chemical Vapour Deposition grown layer 10 of low stress silicon nitride with a thickness of 1 micron. With a suitable mask a photo lacquer pattern with 2 micron orifices at the front side of the wafer 12 and a similar pattern with 15 micron openings at the back side is being exposed and developed. With the aid of anisotropic reactive ion etching a nozzle orifice 11 with a diameter of 2 micron and a length of 1 micron is made in the silicon nitride layer and with use of dry and wet chemical KOH etching a cavity 13 with a diameter of 15 micron and a length of 400 micron is made in the silicon wafer 12.

The flow rate Φ of a medium or a liquid with viscosity η through an orifice (tube) with length L and diameter D for viscous flow at a pressure difference ΔP is given by the law of Poiseuille: $\Phi_{Poiseuille}=\pi D^4 \Delta P/128L\eta$. A parabolic velocity pattern with a low velocity along the wall and a high velocity in the middle of the tube will settle in case the length of the tube L is larger than typical six times the diameter D. The mean velocity v of the medium or liquid is always given by $v=4\Phi/\pi D^2$.

In case the length L is of the order of the diameter D the law of Poiseuille will change to the law of Stokes: $\Phi_{Stokes}=D^3\Delta P/24\eta$. The parabolic velocity pattern will not be valid in this regime. Dagan et al., Chem.Eng.Svi., 38 (1983) 583-596 have proposed an interpolation formula for both regimes: $\Phi_{Dagan}=D^3\Delta P/24\eta\ [1+16L/3\pi D]^{-1}$.

At large velocities v the viscous regime will not be valid any more because another force/pressure is necessary for a kinetic (inertial) contribution to accelerate the medium or fluid to a velocity v. This pressure difference is given by $\Delta P_{kin}=0.5\rho v^2$, with ρ the mass density of the fluid v (cf. Law of Bernoulli).

An important insight according to the invention is that the total needed pressure ΔP_{tot} is the sum of the viscous and the kinetic contribution:

$\Delta P_{tot}=\Delta P_{vis}+\Delta P_{kin} = 6\eta\pi[D+16L/3\pi]^{-1}\ v + 0.5\ \rho\ v^2$.

Typical for a waterbased fluid and for a thin orifice this means:

$\Delta P_{tot} \approx 18.000D^{-1}\ v + 500\ v^2$

(L<D, $\eta=10^{-3}$ poise, $\rho=1000$ kg/m ^{3}, D in micron, v in m/s, ΔP in Pascal).

At a ΔP_{kin} of 4 bar (=4x10^5 Pascal) the maximum jet velocity will be 28 m/s.

ΔP_{vis} will be for this velocity 500.000 D^{-1}. In case D > 2 micron than ΔP_{vis} < 2.5 bar. ΔP_{tot} is then 4 + 2.5 = 6.5 bar, less than the maximum of 10 bar. However in case L/D > 2 then at D= 2 micron the needed pressure will surely exceed 10 bar.

Another important insight is that with a very thin orifice (L≈D≈micron) both in the viscous and in the kinetic regime all the fluid will leave the orifice as a jet with constant velocity v (no parabolic velocity distribution). Especially the kinetic energy of the jet will make that the jet will prolong its track before it breaks up in small droplets, which is particularly useful for Rayleigh break-up of the jet in droplets in air. Rayleigh droplets have a typical droplet size 1.6 times the diameter D of the out coming jet. The fabrication tolerance in the diameter D of the nozzle orifice is an essential factor in determining the amount of liquid ($\Delta V=4\pi\ (1.6D/2)^3/3$) in a Rayleigh droplet. The United States FDA imposes a repeatability of 20% for 90% of the droplets and 25% for the remaining 10%. Only micro machining methods are capable of

producing orifices with a tolerance less than 3% (= variation in $\Delta V < 10\%$). Also because micro machining is done in a sterile and particle free Clean Room environment also the effect of fouling of the nozzles due to particles and/or micro organisms is avoided.

Another important insight according to the invention is that for a very thin orifice($L \approx D \approx$micron) the flow rate at relatively low pressures (3-10 bar) is mainly determined by the kinetic contribution, which means that viscosity of the fluid (medicine) has a minor role as long as $L \approx D$ and $\eta < 10^{-2}$ poise. Jetting (e.g.Rayleigh break-up) with a nozzle plate with a thickness less than 2 micron and orifices with a diameter between 0.4 and 10 micron at a pressure in which the contribution of the kinetic regime ($0.5\rho v^2$.) is larger than the contribution of the viscous regime ($6\eta\pi[D+16L/3\pi]^{-1}.v$) is therefore a very good method to deliver and dose medicines nearly independent of the viscosity of the medicine.

Another important insight according to the invention is that medicine (e.g. proteins and peptides) degradation is strongly diminished if such thin orifices are used at relatively low jetting pressures (<10 bar) with a minimum of shear in strength, time and length of the medicine in passing such an orifice.

Using the law of Stokes and Poiseuille (or Dagan) it is easily to calculate that the flow resistance of the 2 micron orifice 11 is still 5-10 times higher than the flow resistance of the cavity 13 with diameter 15 micron and length 400 micron. This means that the pressure/flow characteristics of this structure is still mainly determined by the 2 micron orifice.

In preference the thickness of the nozzle plate 10 for atomisation is less than six times the diameter of the nozzle orifice 11 and in preference less than one to two times the diameter in order to prevent the built up of a parabolic velocity distribution. The flow resistance may be further reduced through the manufacturing of tapering orifices although it is well known that the amount of tapering is very difficult to control precisely. In case the nozzle plate 10 has a thickness less than 2 micron it still has sufficient strength and it is not necessary to taper the orifices.

Nozzles can be used for as well atomisation and filtration. An embodiment of a nozzle with a nozzle orifice for atomisation or filtration 4 is shown in **FIG. 5A, 5B** and topview **Fig. 6.** The nozzle plate **40** comprises more nozzle orifices **41** placed next to each other in order to increase the throughput. The nozzle plate **40** with a thickness of 1 micron of low stress silicon nitride has a width of less than 250 micron **30** and a length of more than 300 micron **31**. The maximum pressure strength of each nozzle plate **40** is well above 10 bar. A nozzle plate **40** with a width of 100 micron has a pressure strength well above 20 bar. A number of those nozzle plates **40** are closely packed with a mean distance less than 100 micron offering a large effective nozzle plate area as seen in topview **FIG. 6.**
The nozzle **4** comprises further at least one shallow flow channel **44** connected to the nozzle plate **40** with a mean depth of minimum 10 and of maximum 300 micron connected to the nozzle plate. This depth **43** is dependent on the size and number of the nozzle orifices **41** in the nozzle plate **40**. The flow resistance of the flow channel **44** in the nozzle plate support should be at least one to ten times smaller than the flow resistance of the nozzle plate **40** itself. In case the total flow resistance of the nozzle plate support as defined by regions **44** and **45** is one to five times the flow resistance of the nozzle plate **40** a nice flow limitation has been constructed in case the nozzle plate **40** would disrupt. Alternatively two or more openings **46,47** can be provided in each nozzle plate to promote fluid flow and the removal of particles and air bubbles underneath the nozzle plate **40**.
Cross-flow cleaning **90,91** on both sides of the nozzle plate is enhanced by the interconnection **81** in one or more directions of all nozzle plate support flow channels **44** (**FIG. 7A,7B**). Silicon bars **92** between the nozzle plates **40** may be provided for enhanced strength.

Subsequently the nozzle plate **50** may be chosen thicker than a few micron with corresponding tapering orifices **51** in order to reduce the flow resistance still further, shown in **FIG. 8A.** A good measure is also to make spiral grooves **55** in the nozzle orifice **51** to give the medium a rotational motion when leaving the orifice **51**, shown in **FIG. 8B.** Anisotropic and directional etching techniques with SF_6 and O_2 at low biasvoltage 10-40 eV make it

possible to make such grooves in e.g. a <100> silicon wafer. The groove **55** will start at a defined rectangular orifice **52**, the groove will turn and will stop turning as defined by the orientation of the <111> planes **56** shown in **FIG. 8C.**

With preference a number of nozzle orifices **61** are placed very close together (**FIG. 9**), which improves flow rate, filtration and kinetic jetting behaviour, e.g. 2 or more nozzle orifices with a diameter of 2 micron may be separated with a mean distance less than 0.5 micron **62.**

Nozzle plates can be made substantially stronger (up to 250%) when the nearest distance **100** between all nozzle orifices and the nozzle plate support is at least six times the thickness of the nozzle plate **FIG. 10A, 10B.**The pressure strength of the nozzle plate may be further increased with at least 50% when the orifices are placed in a triangular or rectangular pattern **101** with respect to a long side of the nozzle plate support. Preferential the orifices are slit shaped and placed parallel along the width of the nozzle plate support, **FIG. 10B.** An organic coating **104**, in particular a parylene coating on the nozzle plate may further increase the pressure strength of the nozzle plate. Also a bacteria killing surface modification **105** may be applied, for example a silver coating, **FIG. 11.** A silicon nitride coating on the nozzle plate and the nozzle plate support may also be provided to make the whole structure inert for acid and caustic.

A next embodiment of a nozzle for atomisation is shown in **FIG. 12,13** and **14.** The nozzle plate **10** with a thickness of 1 micron comprises circular orifices with a diameter of 0.8 micron. The distance between any of two orifices of the nozzle plate is larger than five times **110** the nozzle diameter in order to prevent recombination of droplets formed of nozzles next to each other. **FIG. 13** shows a cross section of a nozzle plate wherein the orifices are slightly protruding out **130** of the surface (0.1 – 2 micron) of the nozzle plate **10.** This measure is particular useful if the nozzle plate is used for jetting of a spray or emulsification because it prevents that the droplets will adhere to the surface of the nozzle plate just next to the nozzle orifice, resulting in to large droplet formation. In case of atomisation (**FIG. 14**) it is particular useful to make a small area (e.g. 2-5 micron radius around the nozzle) hydrophobic **140** to

prevent droplet attachment/smearing and an unwanted increase of the jet diameter (confinement of the jet). Preferential the next surrounding area **141** should be made hydrophilic in order to drain a too large formed droplet that might inhibit jetting behaviour along the hydrophilic coated part of the nozzle plate. Good results have been obtained with a micro porous hydrophilic coating (PolyVinylPyrolidone/ PolyEtherSulphone) with a thickness of a few micron on area **141**. Alternatively capillary forces may be used to drain the droplet through e.g. the provision of a drain channel **156** with a height of e.g. 2 to 50 micron between the nozzle plate and an e.g. parallel placed drain plate **155** with an orifice **154** larger (e.g. 5 micron) than the jetting orifice (e.g. 1 micron), see **FIG.15**. The jet will normally first pass the jetting orifice and next the larger orifice without fluid contact with the drain plate, but as soon as a droplet is formed that touches the drain plate it will be drained through the drain channel. A hydrophilic inner surface **142** of the nozzles of the nozzle plate is a good measure in order to suppress a large Pascal Pressure with at least 50%.

Jetting may be enhanced by using a piezoelectric actuator at a frequency between 100 kHz and 3 Mhz. Jetting may also be enhanced using the eigenresonance frequencies of the nozzle plate. This frequency should match the value of the initial jet velocity divided by two to two hundred times the diameter of the nozzle, typically a value between 50 kHz en 5 MHz. The eigenresonance frequency is mainly determined by the mass and a fortiori lateral dimensions of the free hanging nozzle plate (typical 1x10x10micron to 4x250x2000 micron), the rigidity of and the tensile pre-stress in the nozzle plate (typical 10^6to10^9 Pascal). A vibrating nozzle plate **150** is shown in **FIG. 15**

Measures to prevent droplet coalescence include: to charge the droplets during droplet formation with an external voltage, or by friction (tribocharging) of the fluid in the nozzle plate device, or by friction of the droplets with the air. An electrical connection (short-circuit) between the patient and the atomising device may be necessary (patient serves as an earth electrode). Further measures include placing the nozzle plate at an angle **160** between 10° and 90° in a cross flow channel **161**, particularly useful for Rayleigh break-up (**FIG. 16**) or co-flow of air or another medium through separate nozzles orifices. Good results have been obtained with a nozzle plate design (**FIG. 17**) in which the nozzles for atomisation are lined up

and the nozzles for co-flow **170** are lined up perpendicular to the cross flow channel. The nozzles for the liquid and the nozzles for the air have separated **172** supply channels. For emulsification it is particularly useful to place the nozzle plate in a dead-end configuration in the channel, which channel **181** narrows significantly at the downstream side. With relatively large nozzle orifices it is then possible to make very small emulsion droplets or gasbubbles(**FIG. 18**) at least 2 –10 times smaller than the diameter of the smallest nozzles **180**. Double emulsions (o/w/o or w/o/w) can advantageously be made with this device, because small emulsion droplets can easily pass the larger orifices without coalescense.

FIG. 19 and **20** show an embodiment of a filter means or nozzle for retaining particles. The nozzle plate support body is made out of one silicon <110> wafer. With this type of crystal orientation it is possible to make perpendicular openings in the support by use of wet (KOH) etching. When the width between two walls of the nozzle plate support body **191** is chosen small (e.g. 20 micron) it is possible to make a high flow resistance in the nozzle cavity. For filtration purposes it has proven to be useful to make an open porosity higher than 30%, not only because of the high flux, but also because it is then relatively easy to back-flush the membrane to prevent irreversible fouling.

The nozzle plate may also be used for retaining and subsequent microscopic observation **192** of these particles, e.g. bacteria's, yeast cell's, blood cell's, etc. Fluorescent dyes may be used to simplify and identify specific species of the micro-organisms on the filter. Silicon nitride and other inorganic nozzle plate materials have the advantage in contrast to many organic polymeric materials that there is virtually no auto-fluorescence signal from the material itself. In some cases it is convenient to place the nozzle orifices further apart, in order to isolate the micro-organisms from each other for a more easy recognition and enumeration.

Very useful nozzle plates for this purpose are characterised in that the spacing **200** between the nozzle orifices is minimum three and maximum thirty times the diameter of the nozzle orifices.

Filter means or nozzles may be used for disposable filtration applications, with preference small nozzle plates 220 (e.g. 5x5mm) are embedded in a ring shaped support 221 (e.g. ABS plastic discs) with outer dimension of e.g. 1.0, 2.5 and 5 cm in diameter and ready to use in standardised commercial filtration holders. With preference the nozzle plates are countersink 222 with a depth of 10 to 500 micron in the ring shaped support to prevent contamination, to facilitate packaging and mechanical rupture of the nozzle plate (FIG. 22).
For reusable application an optic transparent cover slip 230 is placed over the nozzle plate in such a way that a cross-flow channel 231 with a depth of 50 to 500 micron exists between the nozzle plate and the cover plate (FIG. 23). With preference the cover plate is a glass like material that is anodically bonded to the nozzle plate or the nozzle plate support body at elevated temperature (300-400°C) at a voltage between 500 and 1500 V. Cleaning and reuse of this device is facilitated 232 by using ultrasound with a frequency between 100 kHz and 1 MHz. A liquid handling board 234 can be made in glass (with a preferred thickness between 0.5 and 11 mm) for supply of liquid to and from the nozzle plate. By using an anodic bond between the nozzle plate or the nozzle plate support body and the liquid handling board, glass can be used as a liquid handling board for applications in which the required pressures are higher than 0.8 bar.
With preference the nozzle plate support body has cavities 233 with at least the same size as the nozzle plate. It is then possible to use a microscope 192 with a light source that projects light 193 first through the nozzle support and next on the nozzle plate. Most microscopes with phase contrast mode work in this manner. FIG. 28 shows a nozzle plate 340 that can also be used for the deposition (stencilling) 343 of isolated material spots 342 on a substrate 341 with feature sizes determined by the lay out of the nozzle plate. <110> silicon is a good support material for the nozzle plate for these purposes.

Large nozzle plates with an outer circular diameter of e.g. 2, 3, 4, 6 and 8 inches may be used for micro filtration applications like yeast cell filtration and clarification of beer and other beverages. Sterile filtration of milk and other dairy products is also possible with pore sizes between 5 and 0.22 micron. With a pore size of 0.8 micron it has been tested that a log reduction of 5 to 6 of micro-organisms in milk is well achievable in combination with back-

pulse (pulsed permeate flow reversal) technology. Typical flow rates are 1000 –2000 l/m^2/hour at low trans-membrane pressures (0.03 –0.1 bar) with a back-pulse rate of 0.01 – 5 Hz. The flow rate can be strongly increased (4000-20.000 l/ m^2/hour) using ultrasound in a broad frequency spectrum between 100Hz – 1 MHz. Preferably a frequency is used under 15 kHz or above 50 kHz in order to suppress the cavitation forces that might disrupt the nozzle plates between 15 kHz and 50 kHz. The ultrasound inhibits the forming of a dense cake layer just before the nozzle plate. Alternatively the performance for jetting, filtering, foaming and emulsification may be improved by moving the nozzle plate tangential and/or orthogonal to the fluid in contact with the nozzle plate with an actuator with an amplitude of 0.1 to 100 micron and a frequency of 10 Hz - 10 MHz.

In a special embodiment the nozzle plates or nozzle plate support bodies are bonded to a glass plate in which flow channels **270,284** have been made with the use of grinding or powder blasting (**FIG. 24, 25 and 26**). Glass plates of type borosilicate have the advantage that they are very flat, have nearly the same thermal expansion coefficient 4.10^{-6} °C as nozzle plates with a silicon support. Anodic bonding results in a bond inert for acid, caustic and oxydizing chemicals. The flow channels may be used for permeate flow or alternatively for cross-flow. Preferably the flow channels for cross-flow are placed in comb like structures which taper in length and/or in height. The comb structure has the advantage that the total pressure drop over the shallow channel area **285**, the comb teethes **270**, the inlet **278** and outlet **279** (through the glass plate) can be kept low (less than 100mBar), while the cross flow speed at the nozzle plate surface (at the shallow channel area **285**) is yet high enough (more than 0.1m/s) for the enhancement of continuous removal of particles and yeast cells during filtration. The distance **271** between the teethes **270** of each comb are preferably 0.5 – 5 cm, with a depth of 1 – 5 mm, a width of 1 – 5 mm and a length depending on the outer circular diameter. The tapering of the depth **274** is preferably 10° to 40°. The width **273** is preferably tapering 10-40% per cm length of the channel. In particular when powder blasting is used to manufacture the channels in the glass plate, there is a triangular shape of the channels with a relation of the width of the channel and the depth if 1.2. The tapering is meant for a good redistribution of the fluid from the incoming channel to the outgoing channel in such a way that

the pressure distribution along a single tooth of the cross flow channels is homogeneous while the fluid velocity never reaches zero to avoid hygienic failure. The pressure drop over every single tooth is equal by varying the width and the depth of the tooth. The mean cross flow height between the glass plate and the nozzle plate **276** in the shallow channel area is preferably between 0.1 and 1 mm. As well the cross-flow side as the permeate side may be bonded to a glass plate, also one glass plate may be bonded on both sides with a nozzle plate device. With preference the glass plate is being used for a filtration module, where a larger filtration capacity is achieved by placing a number of nozzle plate devices **301** with spacer structures **300, 302** in a stack (plate and frame module with mirror placing of the glass plate **301** and the nozzle plates **303, FIG. 27A, 27B**. The glass plate acts in this module also as tubing for the cross flow inlet **281, 304,** cross flow outlet **282, 305** and permeate collection **280, 306, 307**. Furthermore, the glass plate may contain holes **283** for easy positioning of the glass plate and the spacer structures. Filtration characteristics may also be enhanced by using rotating nozzle plates with respect to the medium in a module. A piezo transducer for ultrasound can be placed on the back side (non powder blasted side) of each glass plate. A typical longitudinal resonance frequency of a glass plate with a thickness of 10 mm is 250 kHz. The ultrasound may be used either for enhancement of the flow rate during filtration or for cleaning of the nozzle plates after or during the filtration cycle. Of course cleaning after filtration with ultrasound is accelerated using proper chemicals (acid/caustic/enzymes etc.). Normal chemical cleaning procedures as used for micro and ultra filtration membranes can herewith be reduced from 1-2 hours back to 10 seconds - 5 minutes. Cross-flow cleaning on both sides of the nozzle plate is enhanced by the interconnection in one or more directions of all nozzle support openings.

Nozzle plates made with a silicon support can be made chemically inert for caustic media by providing a thin LPCVD grown silicon nitride coating with a typical thickness between 0.01 and 1 micron. Other organic and inorganic coatings like e.g. Al_2O_3, TiO_2, ZrO2, ZrO2/Si_3N_4 may be applied to alter the Zeta potential and/or the wetting properties of the nozzle plate to improve filtration characteristics. Other coatings may also be applied to promote anti-fouling like TiO_2, PTFE, self assembling monolayers (SAM, e.g. based on nitryls, disulfides or thiols) or long polymer chains (e.g. polyethyleneglycol) coupled with an end- or side- group to the

nozzleplate. Dense sol/gel coatings or gas permeation layers like Pd, PdAg may also be applied over and in the nozzle orifices to make ultrafiltration and gas filtration membranes.
An important insight according to the invention is that the combination of nozzle plates, back-pulse technology and ultrasound has proven to be very powerful for the enhancement of flow rate and the prevention of irreversible fouling. Without ultrasound a typical clarification run for beer is 4-8 hours, with ultrasound dosed at intervals of 10 minutes for a few seconds the run can be extended to 4-8 days without the need of chemical cleaning procedures.
Backpulsing for a very short time 10-50 ms at regular intervals 0.01 – 5 Hz during cross-flow filtration at low trans-membrane pressure will lift the cake layer from the nozzle plate and will inject it higher in the cross flow channel where the fluid velocity is sufficient high to take it further away.

Backpulsers are also very suitable to use for up-concentration of samples for the detection and counting of food spoiling or pathogenic micro-organisms, e.g. lacto bacillus, E-coli and legionella. After the up-concentration all micro-organisms are present on the nozzle plate and can be processed for e.g. microscopic observation and PCR amplification. Small nozzle plates of e.g. 4x4 mm can be put easily with a clean and sterile pincer in a small PCR-cup. The nozzle plate can also be provided with an immuno binding (or elisa coupling) agent for the selective binding of certain species direct to the nozzle plate during filtration, especially when cross-flow techniques are used for up-concentration of the sample. Magnetic layers may also be deposited for the attraction of immuno magnetic beads. Metallic layers may also be provided on the nozzle plates for e.g. optic non-transparancy, non quenching or electrolysis applications, improvement of filtration under the applicance of a small voltage difference between the fluid and the nozzle plate, or the annihilation(electroporation) of microorganisms under the applicance of a high voltage pulse . Platina may be deposited in electrical resistor strips on the nozzle plate for heating purposes. Also a bacteria killing surface modification may be applied, for example a silver coating. Piezo materials may also be applied for direct vibration of the nozzle plates or for the detection of bending of the nozzle plates for pressure registration. The intensity and the frequency of the backpulsers may also be regulated by the

registration of the nozzle plate trans membrane pressure. The trans membrane pressure will normally increase if there is a built up of a cake layer for the nozzle plate.

Nozzle plates can be made in various ways according to the invention.

A reinforced micromachined polymeric nozzle plate is made by
- depositing a first layer of a photosensitive material, for example negative resist polyimide (Durimide 7510) on a flat and smooth substrate
- exposing the first layer to a suitable light source through a mask (or a laser interference pattern) with a nozzle pattern
- developing and if necessary curing the first layer
- depositing a second layer of a photosensitive material onto the first layer
- exposing the second layer to a suitable light source through a mask with a nozzle support structure
- developing and if necessary curing the second layer
- releasing the thus obtained nozzle plate from the substrate

Another method of making a micromachined polymeric nozzle plate, comprises the following steps
- depositing a first layer of a photosensitive material on a flat and smooth substrate
- exposing the first layer to a suitable light source through a mask (or laser interference) with a nozzle pattern
- developing the first layer
- etching anisotropically the nozzle pattern to a certain depth, typically 1 to 5 micron, in the substrate
- depositing a second layer of a photosensitive material onto the substrate
- exposing the second layer to a suitable light source through a mask with a nozzle support structure
- developing the second layer

- etching anisotropically the nozzle support structure to a certain depth, typically 5 to 500 micron, in the substrate
- electroforming a master mould from the substrate if necessary or using the substrate itself as a master mould
- if necessary depositing a release agent (teflon) on the master mould
- placing a thin sheet of thermoplastic polymer with a typical thickness between 5 and 50 micron onto the master mould

placing a second (flat) substrate with a release agent on the polymeric sheet
- pressing the two substrates to each other with a substantial load at a temperature well above the glass transition temperature of the polymeric sheet if necessary under reduced atmospheric conditions for a short period
- releasing the thus formed polymeric nozzle plate from the substrates at a temperature well below the glass transition temperature

A reinforced micromachined electroformed nozzle plate is made by
- depositing a conductive layer on a flat and smooth electrically insulating substrate
- depositing a first layer of a photosensitive material on the conductive layer
- exposing the first layer to a suitable light source through a mask with a nozzle support pattern
- developing the first layer
- etching the conductive layer with a suitable chemical etchant
- removing the first layer
- depositing a second layer of a photosensitive material with a thickness of at least 2 micron onto the substrate
- exposing the second layer to a suitable light source through a mask with a nozzle device
- developing the second layer such that the remaining resist layer is not in contact with the conductive layer
- putting the substrate in a suitable electroforming bath using the conductive layer as a cathode
- stopping the electroforming process as soon as the electroformed layer has reached substantially at least one or more parts of the remaining resist layer
- releasing the thus electroformed nozzleplate

Another method of making a micromachined nozzle plate device comprises the following steps

- depositing a first layer of a photosensitive material on a flat and smooth substrate, said substrate being covered at both sides with a thin membrane layer
- exposing the first layer to a suitable light source through a mask with a nozzle support pattern
- developing the first layer
- etching the nozzle support pattern in the membrane layer on one side of the substrate and further
- etching chemically the nozzle support pattern through the substrate stopping at a distance of 5 to 100 micron of the membrane layer at the other side of the substrate
- depositing a second layer of a photosensitive material onto the other membrane layer of the substrate
- exposing the second layer to a suitable light source through a mask (or laserinterference) with a nozzle plate structure
- developing the second layer
- etching the nozzle plate structure in the membrane layer
- etching through the nozzles part of the nozzle support structure such that the nearest distance between the nozzles and the nozzle support structure is at least twice the nozzle diameter

Nozzle plates according to the invention may also be used for the extrusion of very viscous media like macromolecular solutions, gel-like solutions and protein-rich media, and for microstructuring of food and pharmaceutical products like e.g. synthetic meat (fibres). Nozzle plates according to the invention may also used for micro-array and micro-titration applications, to make double emulsions and to apply them in bio-capsules because of the small diffusion length of the short nozzle orifice.

Claims:

1. A nozzle device having a nozzle for atomisation of a fluid, the nozzle comprising a nozzle plate support body having a cavity extending from a first main surface to a second main surface thereof, and comprising a nozzle plate having at least one nozzle orifice in fluid communication with said cavity at said first main surface side of said nozzle plate support body characterized in that said support body is provided with filtration means which comprise a filtration plate which is in fluid communication with said cavity at said second main surface side of said nozzle plate support body.

2. A nozzle device as claimed in claim 1 characterized in that the filtration means comprise a filtration plate support body having a cavity which carries the filtration plate at a main surface thereof across from the nozzle plate.

3. A nozzle device according to claim 2 characterized in that the filtration means are directly connected to the nozzle plate support body.

4. A nozzle device according to anyone of claims 1-3, characterised in that the nozzle plate comprises a deepened region stretching towards the nozzle plate support body and in that the at least one nozzle orifice is provided within said deepened region.

5. A nozzle device according to 4 characterized in that said deepened region lies typical 2-200 micron offset to a surrounding portion of said nozzle plate.

6. A nozzle device according to any of the preceding claims characterized in that the filtration plate is provided on the nozzle plate support body at the area surrounding the cavity.

7. A nozzle device according to any of the preceding claims characterized in that the nozzle plate support body is formed of silicon, particularly a <110> wafer.

8. A nozzle device according to any of the preceding claims characterized in that the thickness of the nozzle plate is less than 2 micron.

9. A nozzle device according to any of the preceding claims characterized in that said at least one orifice has a length which is less than six times a diameter thereof, and in particular is shorter than said diameter.

10. A nozzle device according to any of the preceding claims characterized in that said orifice has a diameter between 0.4 and 10 micron.

11. A nozzle device according to any of the preceding claims characterized in that a first zone of said first main surface of said nozzle plate which surrounds said nozzle orifice at least partly is substantially hydrophobic.

12. A nozzle device according to claim 11 characterized in that a second zone of said first main surface of said nozzle plate which surrounds said first zone at least partly is substantially hydrophilic.

13. A nozzle device according to any of the preceding claims characterized in that a drain plate with at least one drain orifice is provided at a distance to said first main surface of said nozzle plate, defining a drain channel.

14. A nozzle device according to any of claims 1-10 characterized in that an area of said first main surface of the nozzle plate at least partly surrounding said at least one nozzle orifice is substantially hydrophilic and that a next area on the nozzle plate, across from said orifice, is hydrophobic.

15. A nozzle device according to any of the preceding claims characterized in that an inner wall surface of said at least one nozzle orifice is substantially hydrophilic.

16. A nozzle device according to any of the preceding claims characterized in that said at least one nozzle orifice slightly protrudes out of said first main surface of the nozzle plate.

17. A nozzle device according to any of the preceding claims characterized in that a first part of the nozzle plate is movable with respect to another part of the nozzle plate, at least at elevated pressures, through the provision of a long slit shaped nozzle orifice.

18. A nozzle device according to any of the preceding claims characterized in that the nozzle plate has been placed at an angle between 10° and 90° with respect to an external flow channel of the nozzle device.

19. A nozzle device according to anyone of the preceding claims characterized in that the nozzle plate is received in flow channel of a flow guiding device, which flow channel tapers down in a downstream direction at least locally.

20. A nozzle device according to any of the preceding claims characterized in that the nozzle plate is provided with a number of orifices for liquid flow together with a number of orifices for gas flow, particularly air flow.

21. A nozzle of the type as applied in the nozzle device of any of the preceding claims for atomisation or filtration.

22. A nozzle according to claim 21 characterized in that at said first main surface said cavity has a cross-section having a width of less than 250 micron and having a length of more than 300 micron

23. A nozzle according to claim 22 characterized in that said width of said cross-section is less than 100 micron

24. A nozzle according to any of claims 21-23 characterized in that the nozzle plate and the nozzle plate support body are covered by a caustic resistant coating, particularly a silicon nitride coating.

25. A nozzle according to any of claims 21-24 characterized in that said cavity within said nozzle plate support body is provided with a relatively shallow flow channel at the first main surface, particularly having a depth of 10–300 micron.

26. A nozzle according to any of claims 21-25 characterized in that the at least one orifice is slit-shaped and placed parallel to a width of the nozzle plate.

27. A nozzle according to any of claims 21-26 characterized in that at the nozzle plate is in open communication with at least one further nozzle plate in one or more directions through at least one additional cavity in the nozzle plate support body.

28. A nozzle according to any of claims 21-27 characterized in that a coating for improved strength is provided on the nozzle plate, particularly a parylene coating.

29. A nozzle according to any of claims 21-28 characterized in that a glass substrate is anodically bonded to the nozzle plate and nozzle plate support body assembly and that the glass substrate is provided with at least one flow channel at a surface thereof which is in open communication with the cavity of said support body.

30. A nozzle according to any of claims 21-29 characterized in that said flow channel is formed by powder blasting.

31. A nozzle according to claim 29 or 30 characterized in that the glass substrate is provided with a number of flow channels wich are placed in comb like structures tapering in length and/or in height.

32. A nozzle according to any of claims 21-31 characterized in that the nozzle plate is provided with a piezo-electric actuator device.

33. A nozzle according to any of claims 21-32 characterized in that the at least one orifice has a groove like structure.

34. A nozzle according to any of claims 21-33 characterized in that the nozzle plate comprises a group of nozzle orifices which are placed closely together.

35. A nozzle according to any of claims 21-34 characterized in that the nozzle plate comprises a zone along a boundary of the cavity which is at least substantially free of any nozzle orifice and has a width which is at least a number of times as large as the thickness of the nozzle plate.

36. A nozzle according to any of claims 21-35 characterized in that the nozzle plate has a porosity of at least 30%.

37. A nozzle according to any of claims 21-36 characterized in that a spacing between said at least one nozzle orifice and a further nozzle orifice is between three and thirty times a diameter of the nozzle orifice.

38. A nozzle according to any of claims 21-37 characterized in that the nozzle plate is embedded in a ring shaped support wich is ready to use in standardised commercial filtration holders.

39. A nozzle according to any of claims 38 characterized in that the nozzle plate is countersunk to a depth of 10 to 500 micron in the ring shaped support.

40. A nozzle according to any of claims 21-39 characterized in that the cavity in the nozzle plate support body exposes at least substantially the entire active portion of the nozzle plate which spans the cavity to enable full microscopic observation.

41. A nozzle according to any of claims 21-40 characterized in that an optic transparent cover slip is placed over the nozzle plate in such a way that a flow channel with a depth of 50 to 500 micron is present between the nozzle plate and the cover plate.

42. A nozzle according to any of claims 21-41 characterized in that the nozzle plate is provided with an immuno binding (or Elisa coupling) agent.

43. A nozzle according to any of claims 42 characterized in that the nozzle plate is provided with a magnetic layer enabling the coupling of immuno magnetic beads.

44. A nozzle according to any of claims 21-43 characterized in that the nozzle plate is provided with a metallic layer facilitating optic non-transparency, non quenching, electrolysis and electric heating applications.

45. A nozzle according to any of claims 21-44 characterized in that the nozzle plate is provided with a sol/gel ultra-filtration coating or a gas permeation layer comprising palladium.

46. Method for micro filtration of beer, milk and other beverages with a nozzle as claimed in anyone of claims 20-44 using ultrasound in a broad frequency spectrum between 100Hz – 1 MHz, preferably under 15 kHz or above 50 kHz .

47. Method of using a nozzle as claimed in anyone of claims 21-46 for jetting, filtering, foaming and emulsification characterized by moving the nozzle tangential and/or orthogonal with respect to the fluid in contact with the nozzle plate.

48. Method according to claim 47 characterized in that the nozzle is moved by means of an electronic actuator which is driven to an amplitude of 1 to 100 micron and a frequency of 10 Hz - 10 MHz.

49. Method according to claim 48 characterized in that the nozzle is freely suspended to alllow vibration at its eigen-resonance frequency.

50. Method according to claim 49 characterized by providing an electrical connection between the nozzle plate and an outer housing of an atomisation device to faciltate a short-circuit between an user and the atomisation device.

51. Method of atomisation of a fluid with a nozzle plate as claimed in anyone of claims 21-46 in which the contribution of the kinetic regime is larger than the contribution of the viscous regime.

52. Method of emulsification with a nozzle plate as claimed in anyone of claims 21-46 with relatively large orifices with a diameter between 1.0 and 50 micron, using an external flow guiding device having a flow channel that narrows down in the downstream direction, at least locally to cover the nozzle plate at least at an active area thereof.

53. Method to make double emulsions with a nozzle as claimed in anyone of claims 20-46.

54. Method of making small shadow patterns on a substrate using a nozzle as claimed in anyone of claims 20-46.

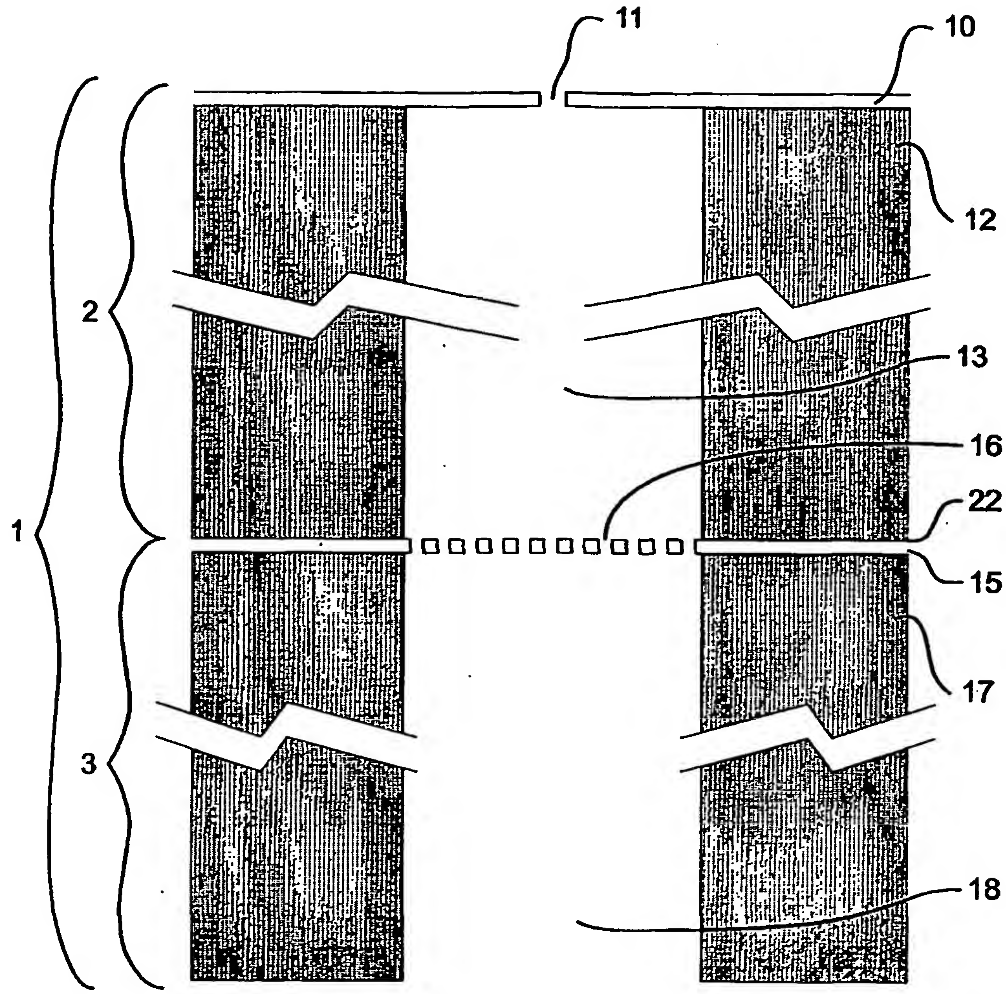


Fig. 1

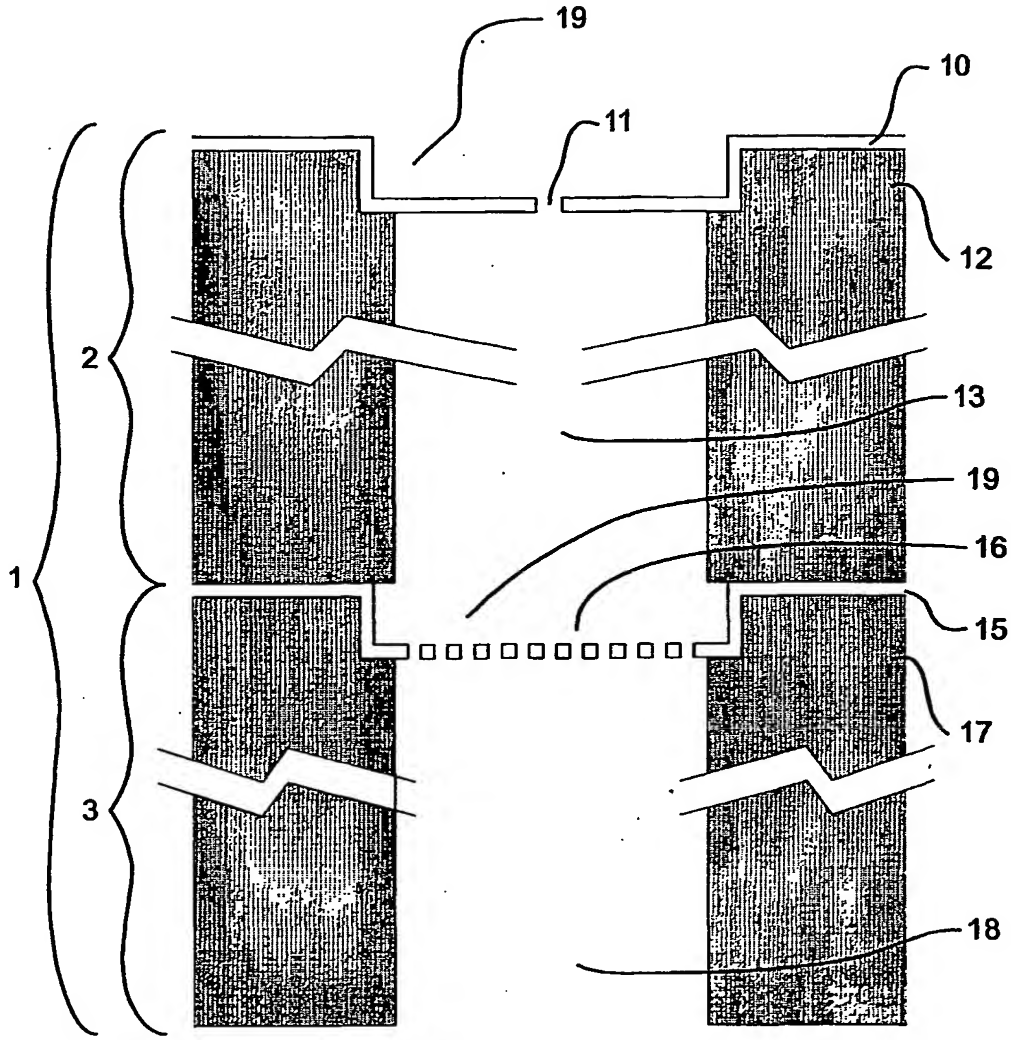


Fig. 2

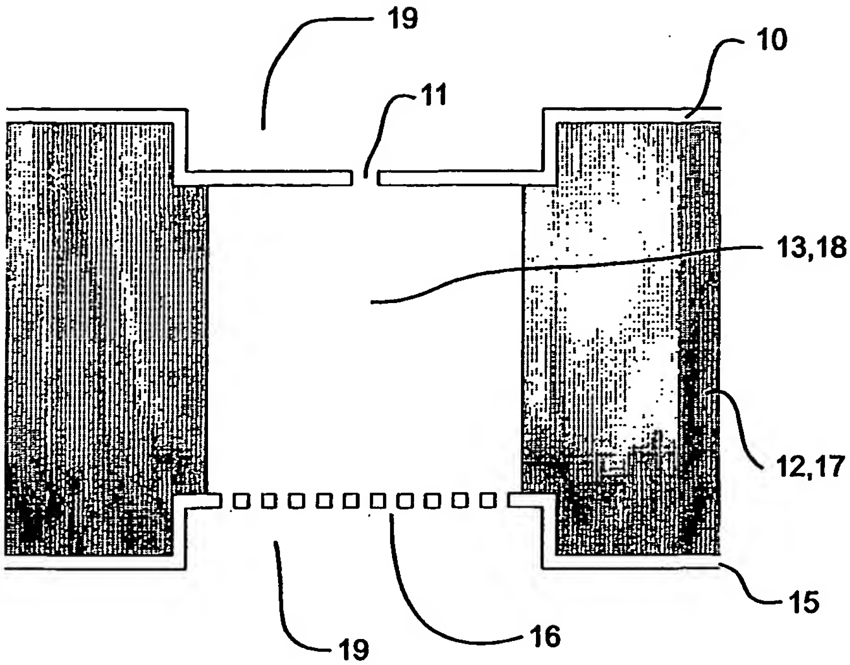


Fig. 3

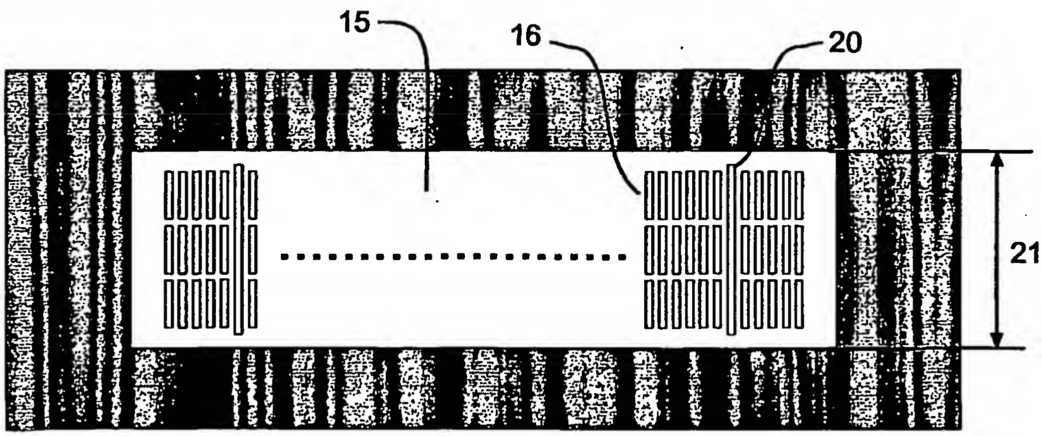


Fig. 4

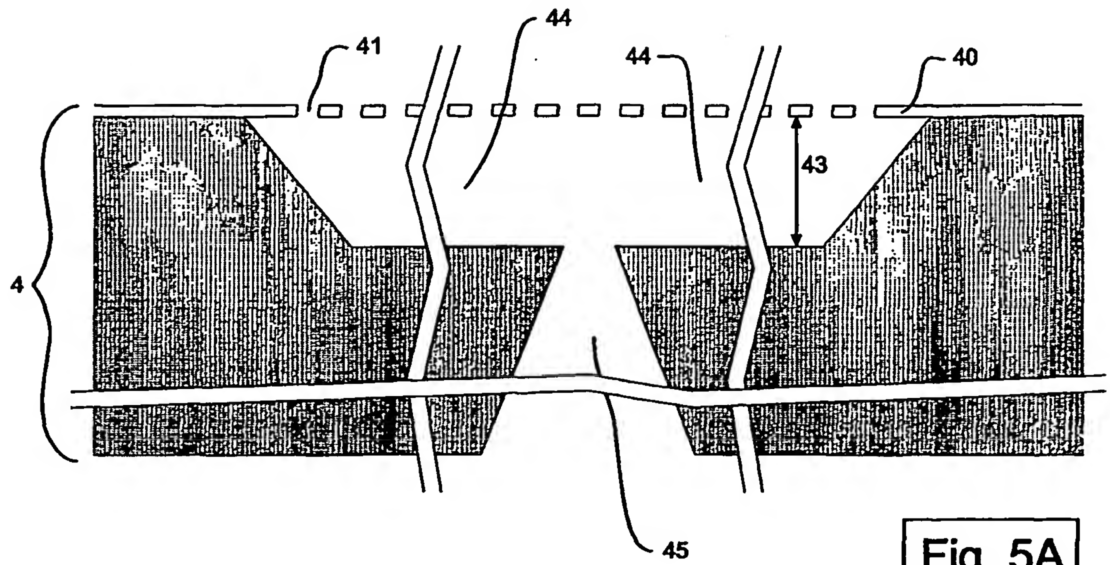


Fig. 5A

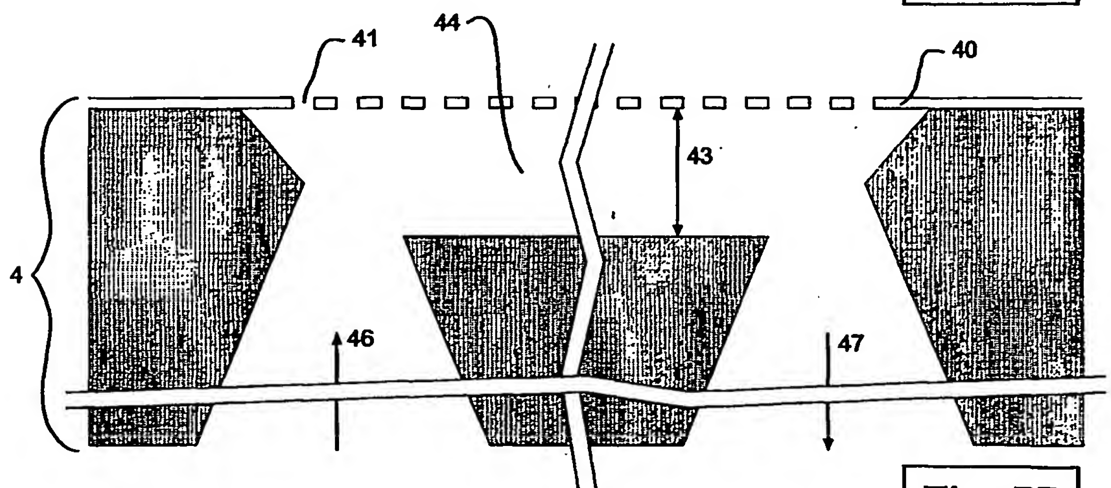


Fig. 5B

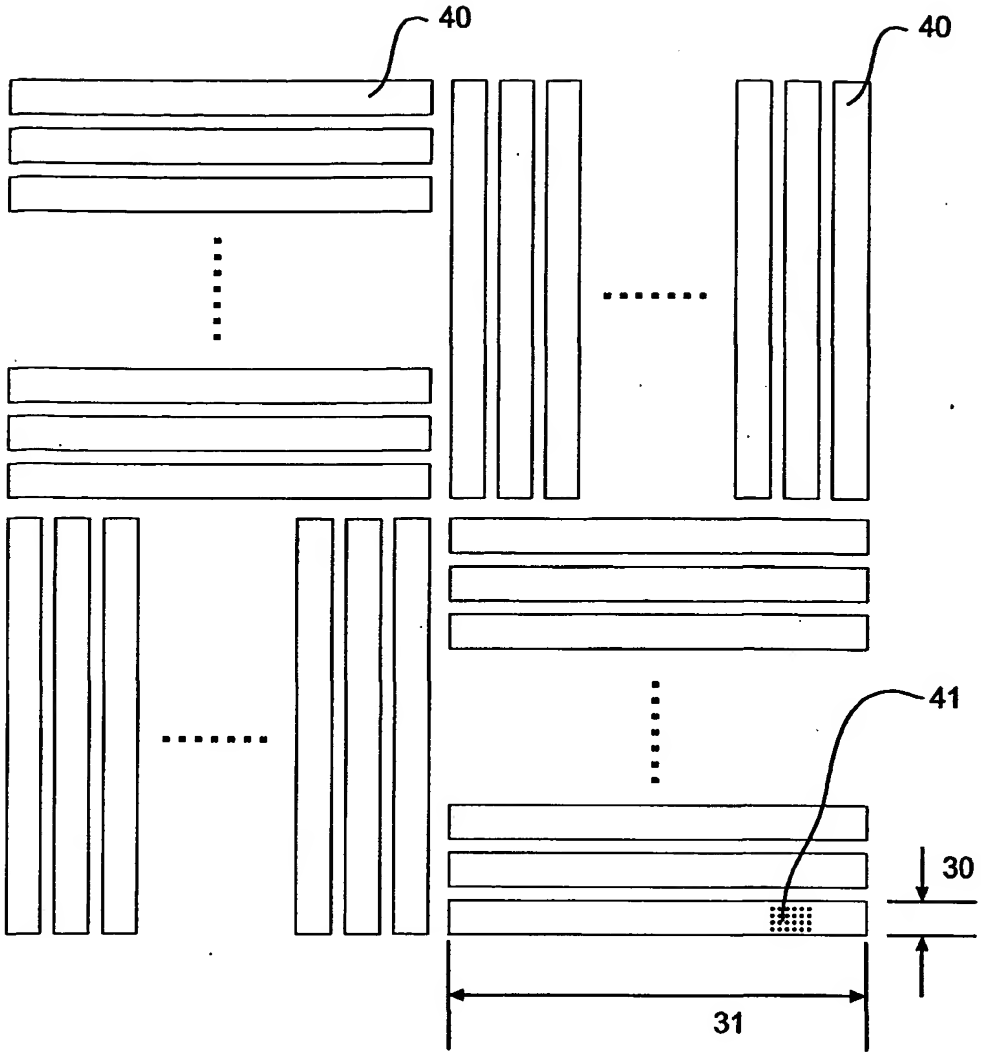


Fig. 6

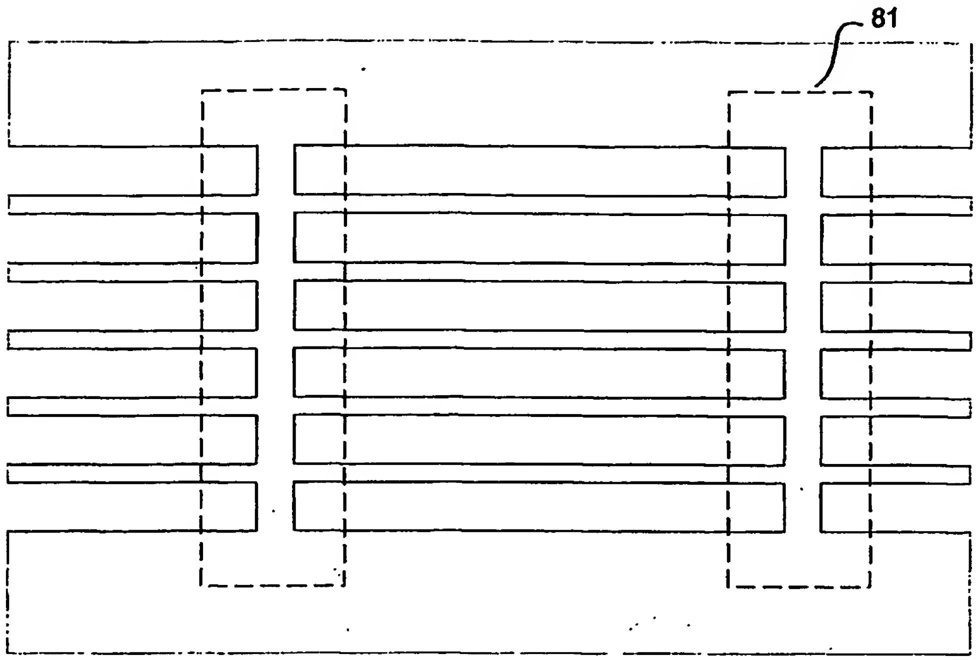


Fig. 7A

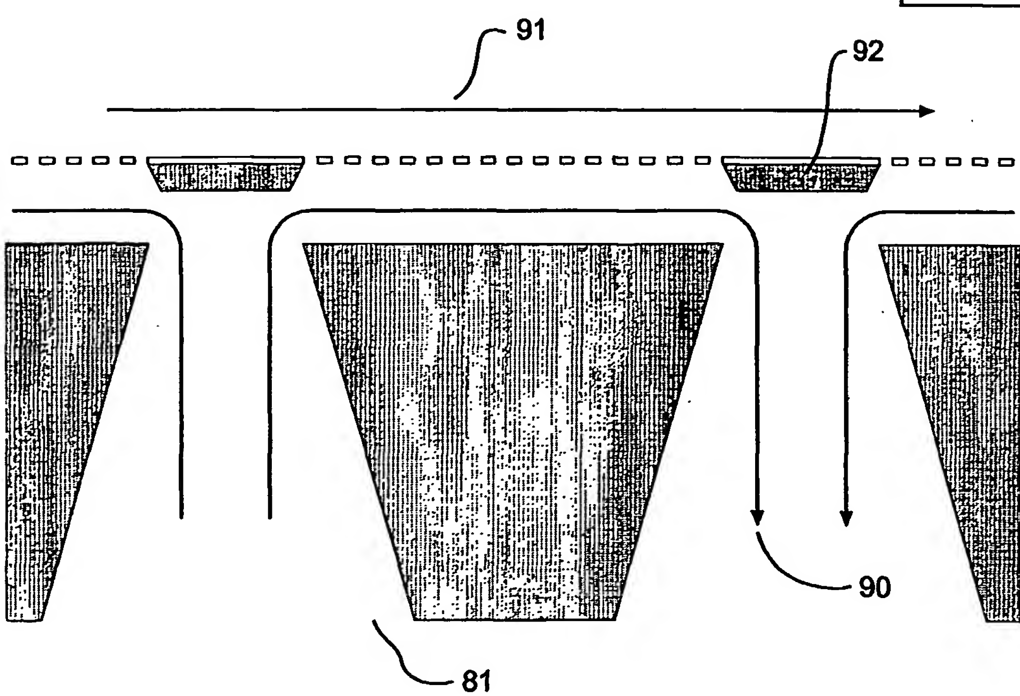


Fig. 7B

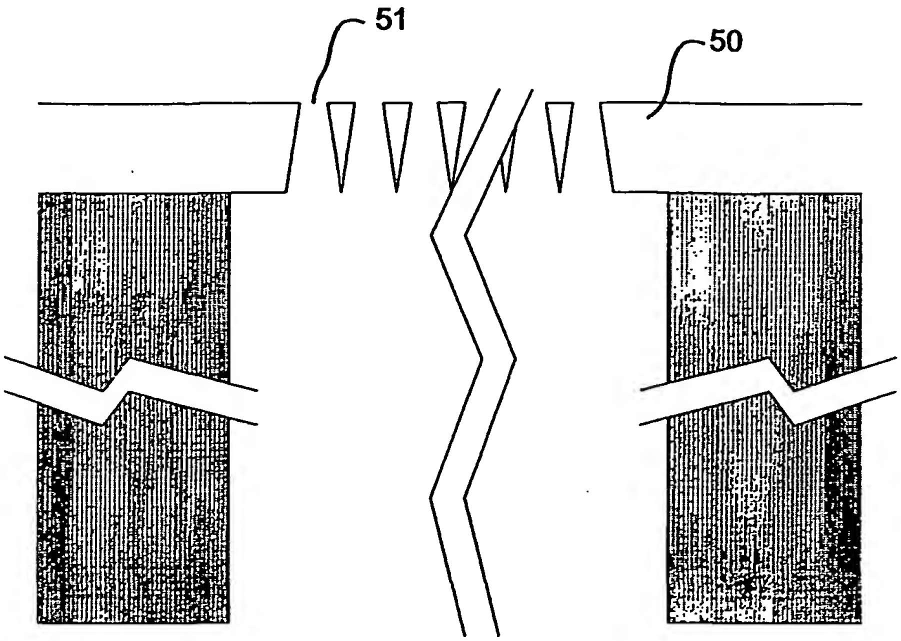
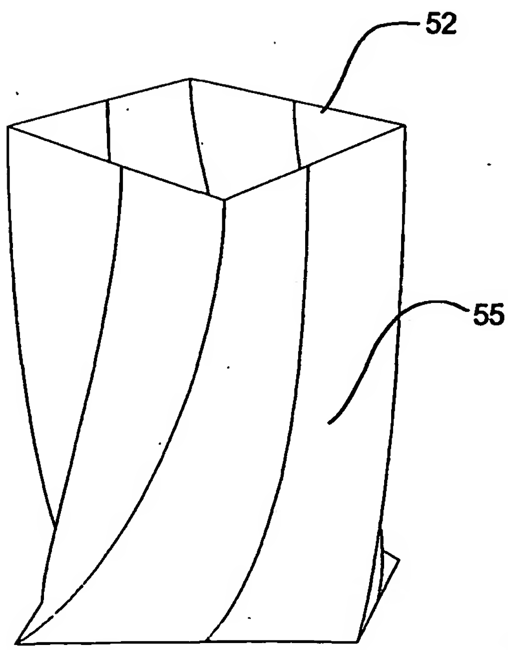


Fig. 8A



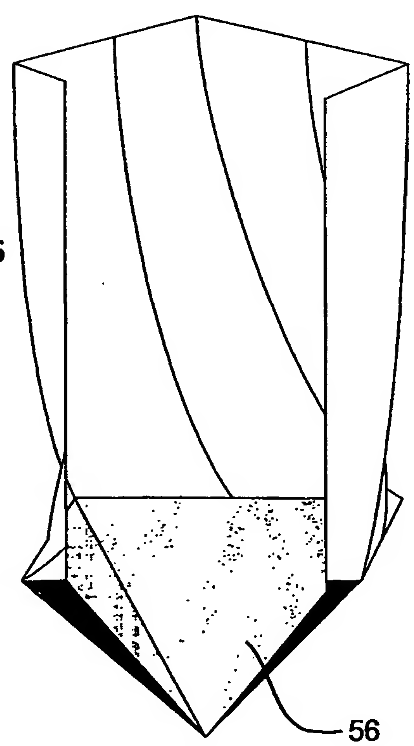


Fig. 8B

Fig. 8C

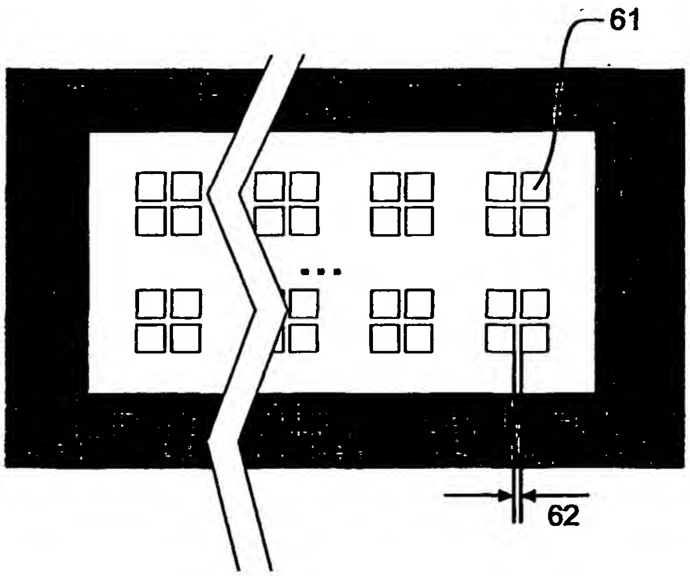


Fig. 9

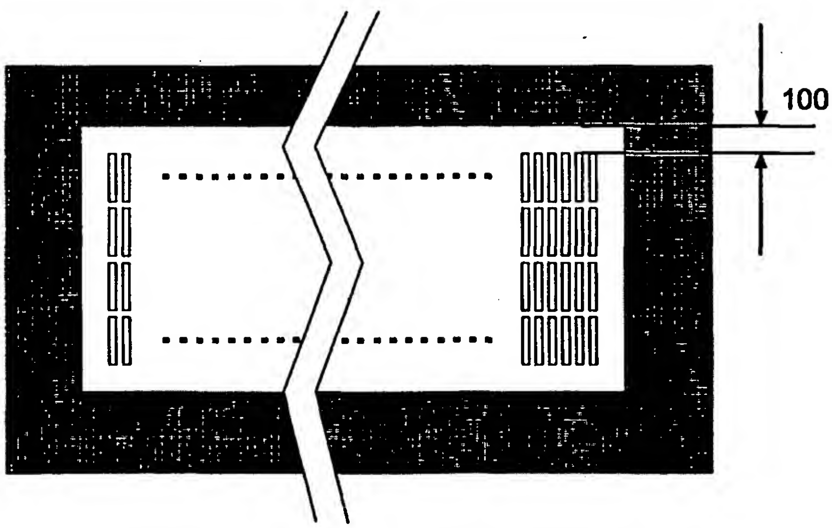


Fig. 10A

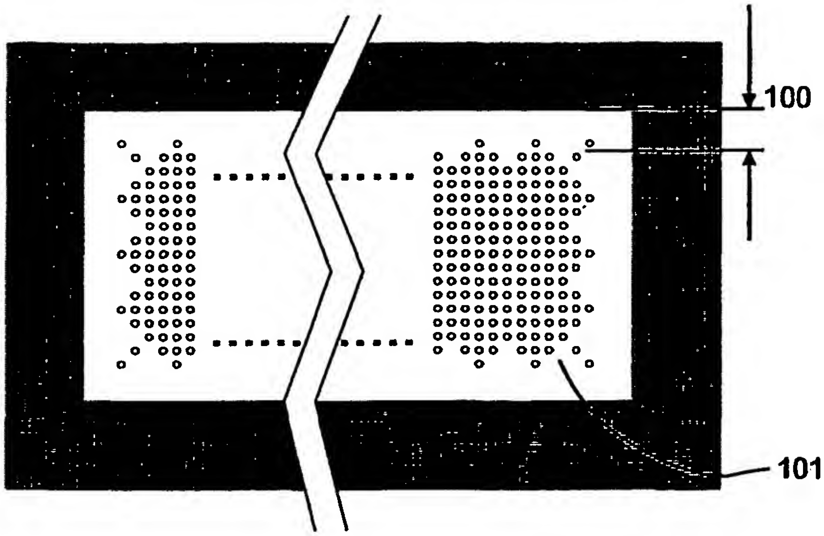


Fig. 10B

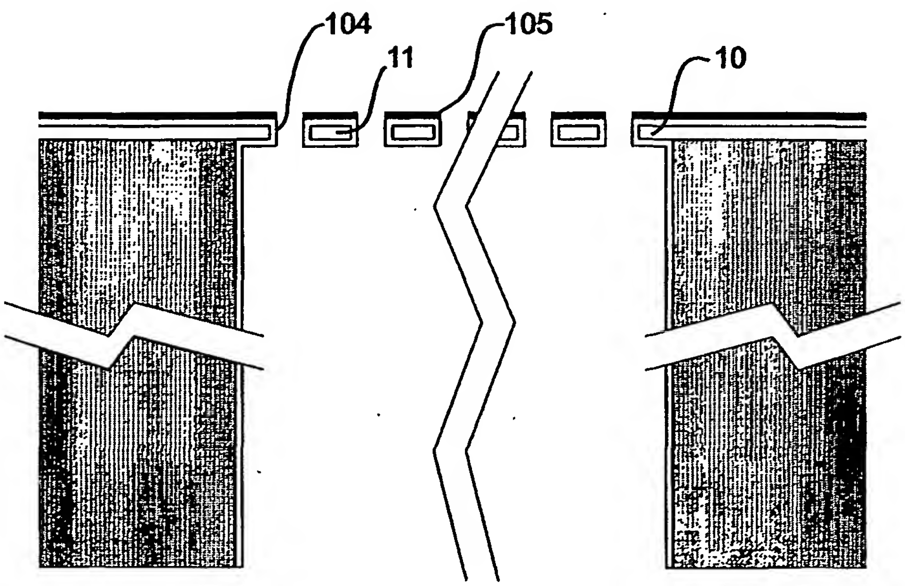


Fig. 11

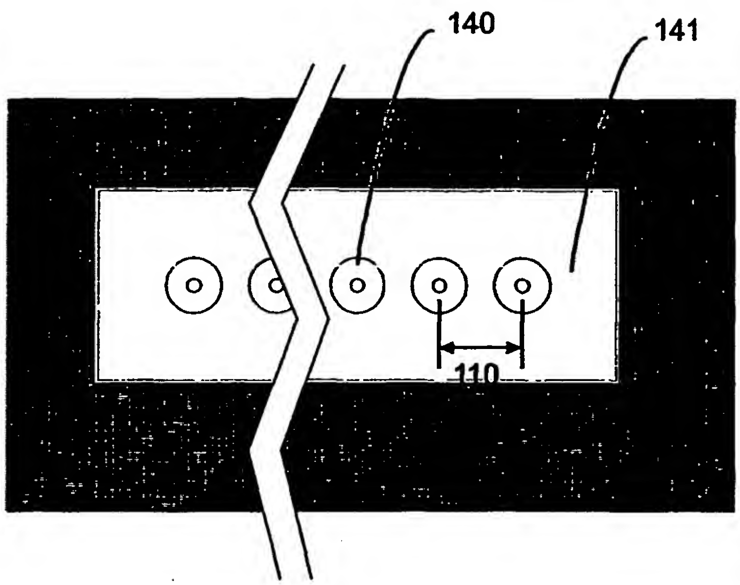


Fig. 12

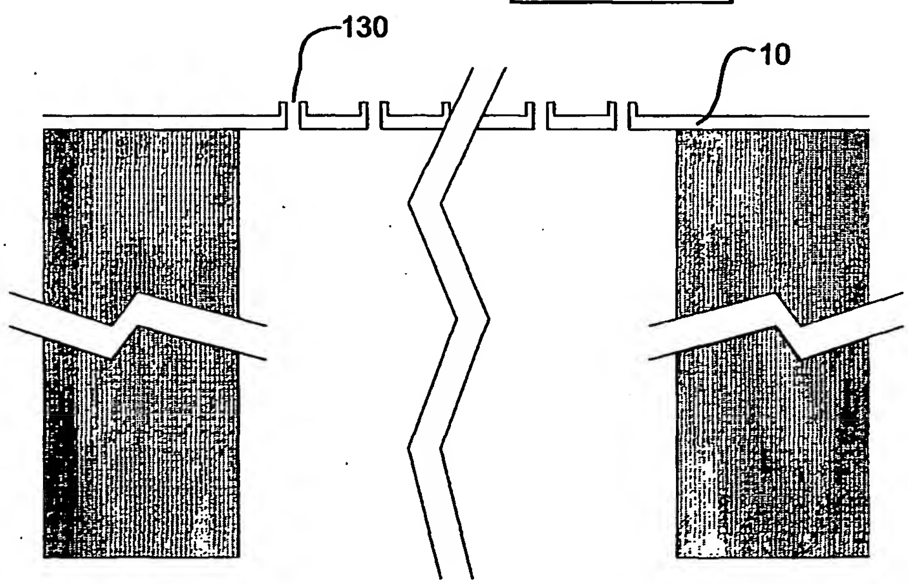
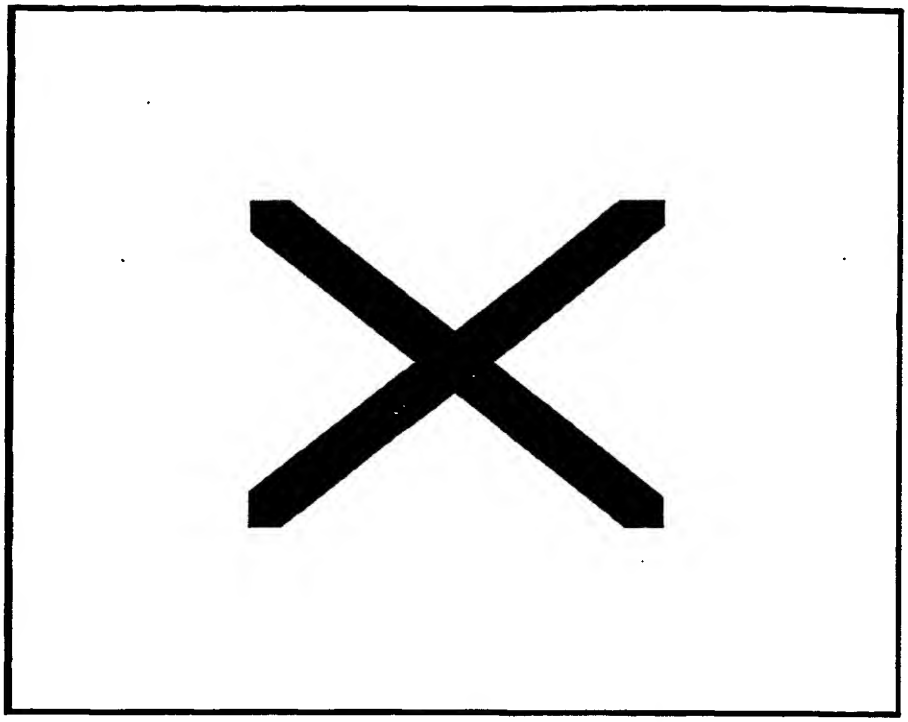


Fig. 13



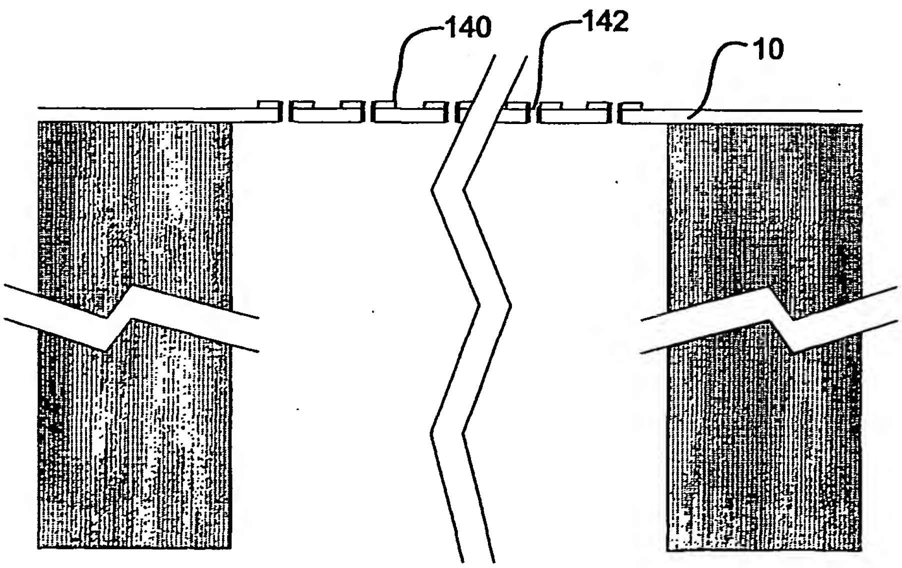


Fig. 14

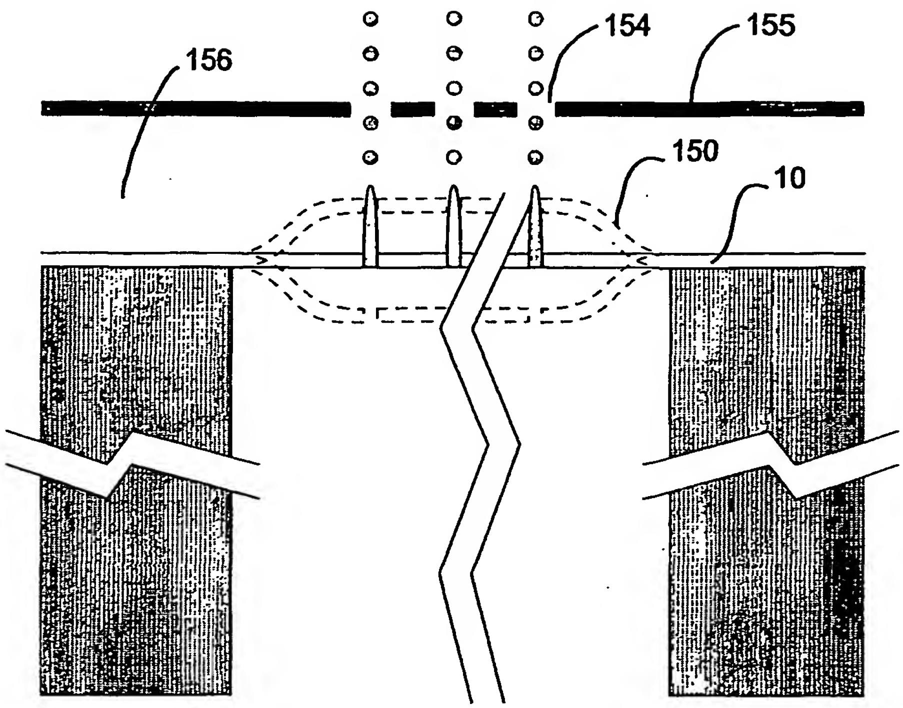


Fig. 15

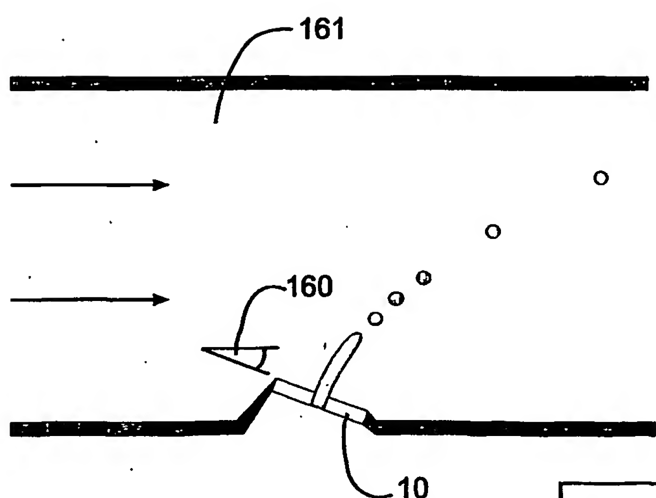


Fig. 16

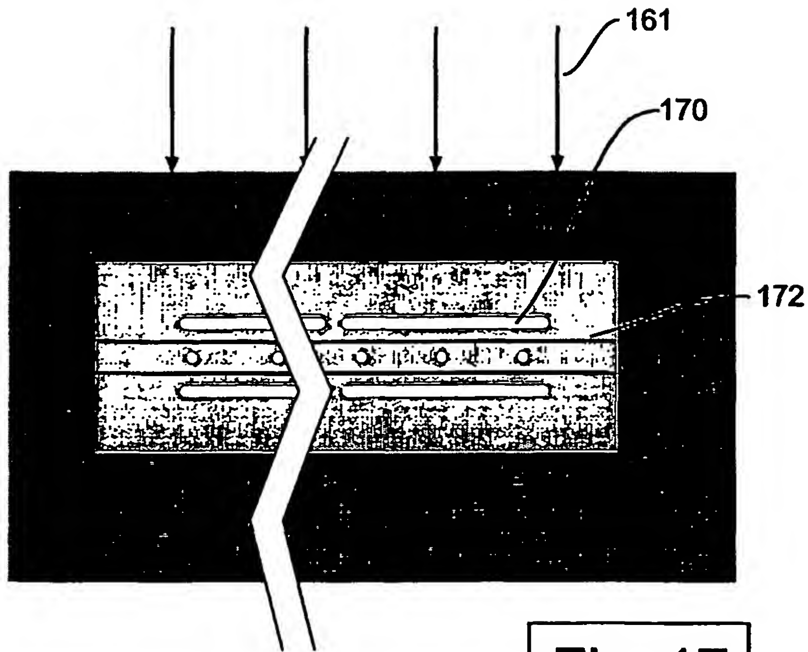


Fig. 17

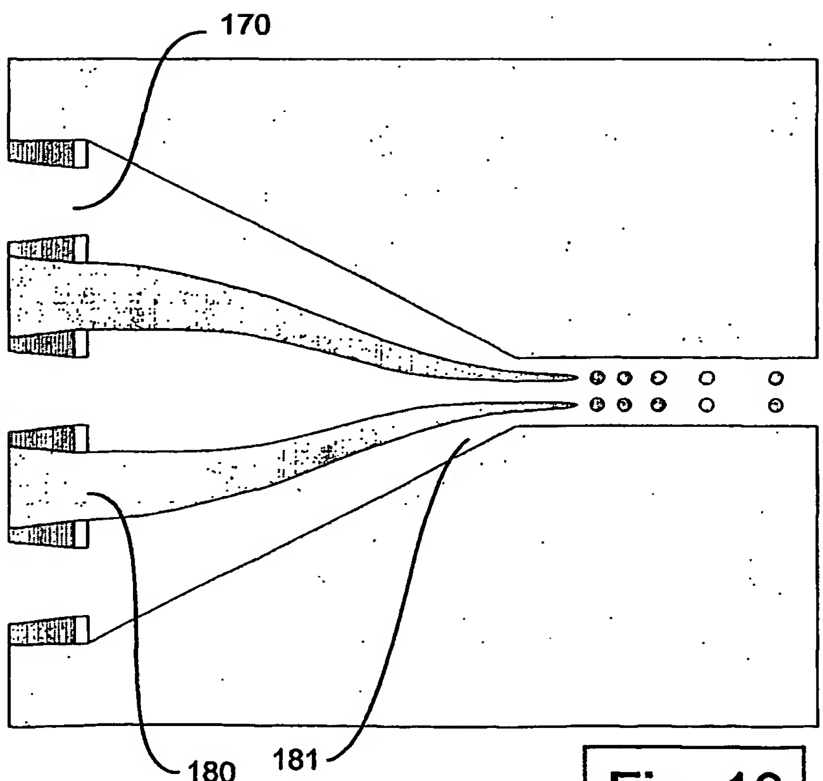


Fig. 18

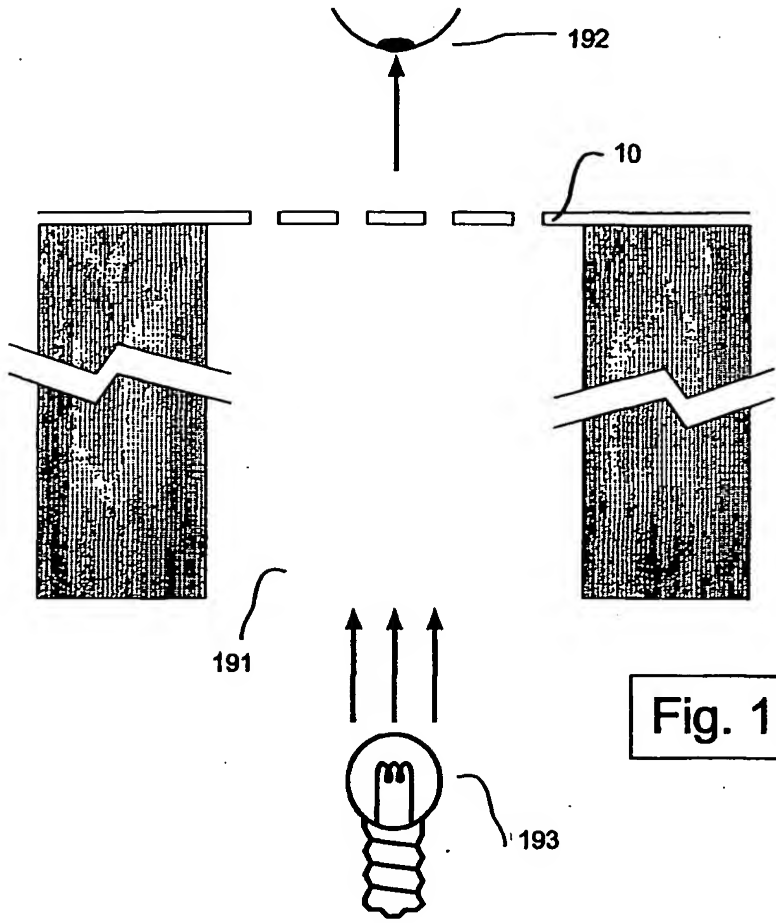


Fig. 19

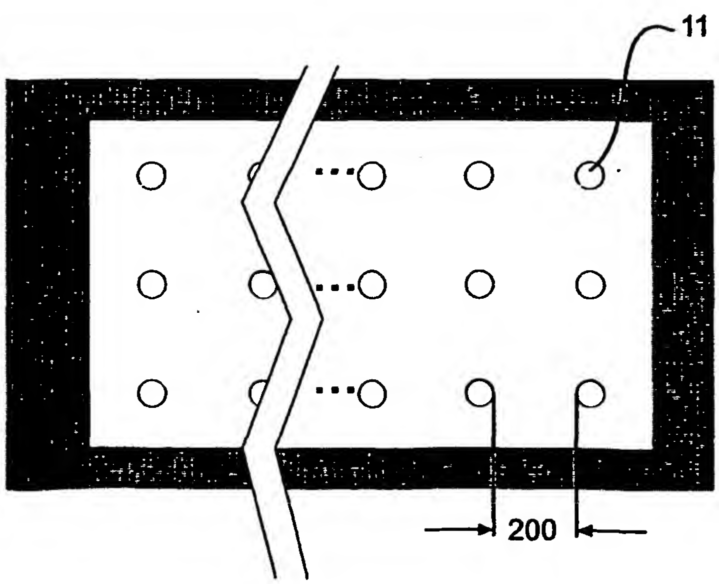


Fig. 20

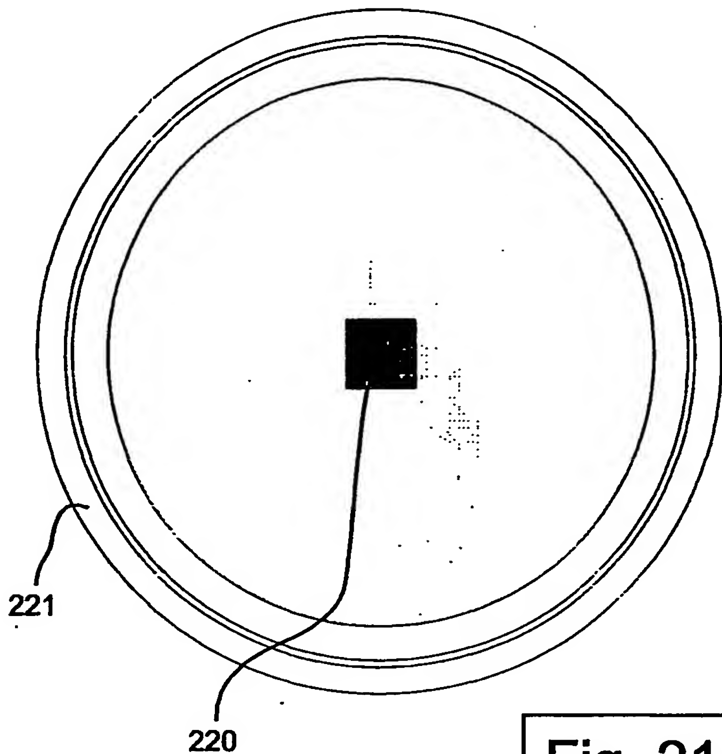


Fig. 21

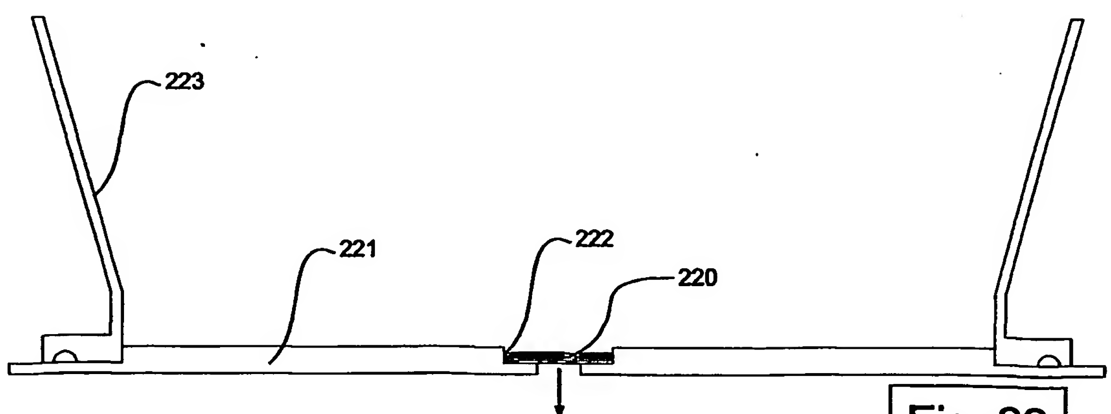


Fig. 22

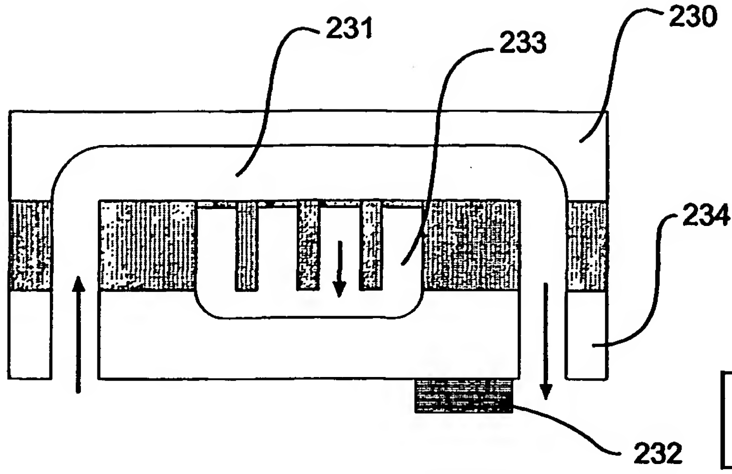

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Fig. 23

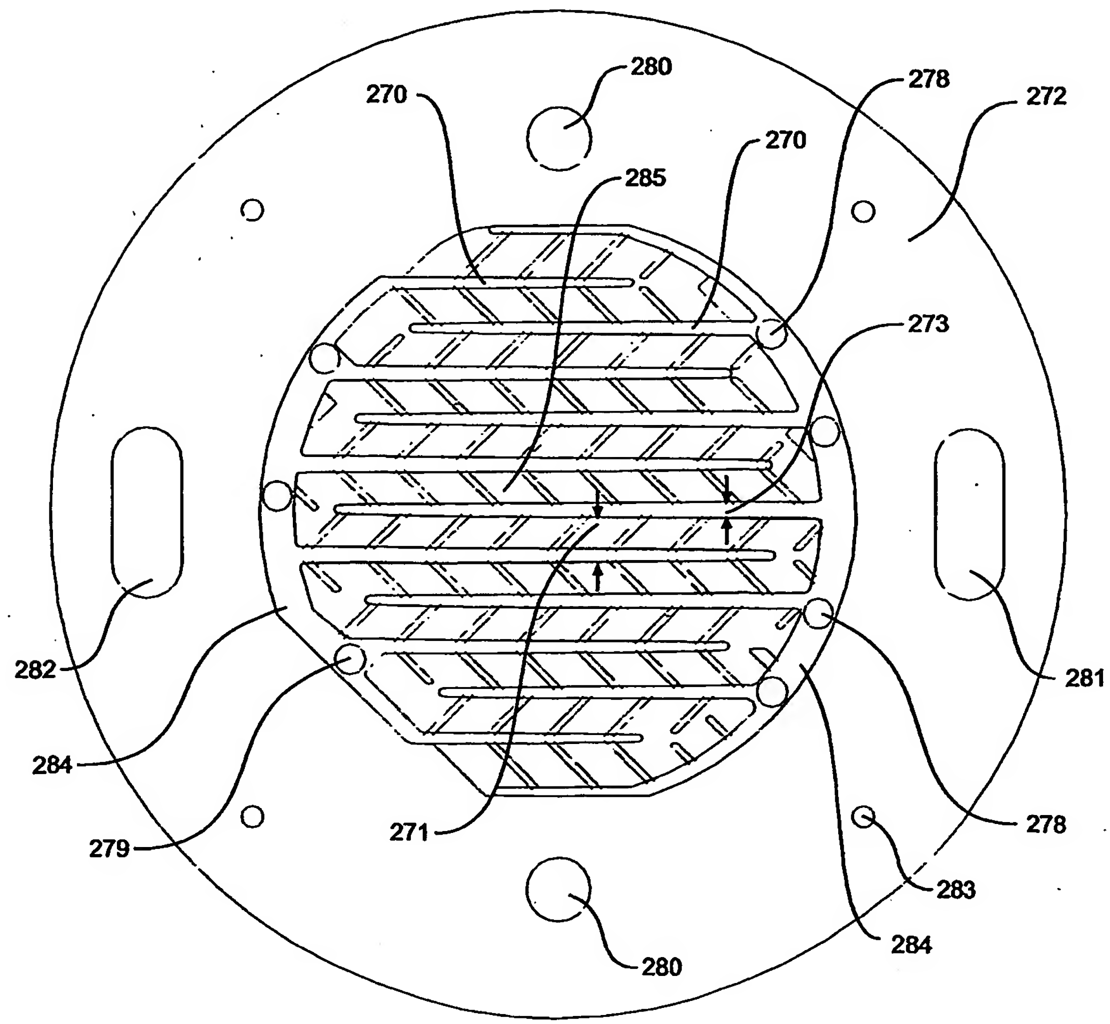
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Fig. 24

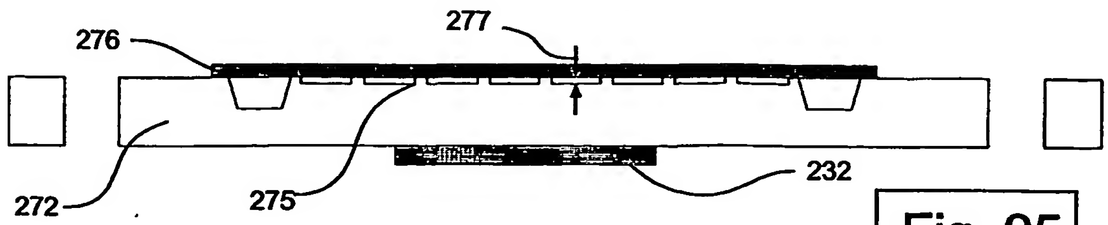

Fig. 25

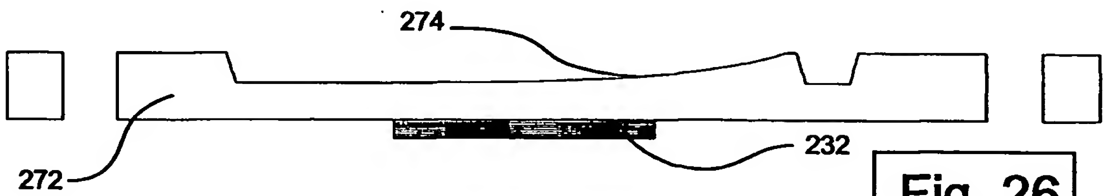

Fig. 26

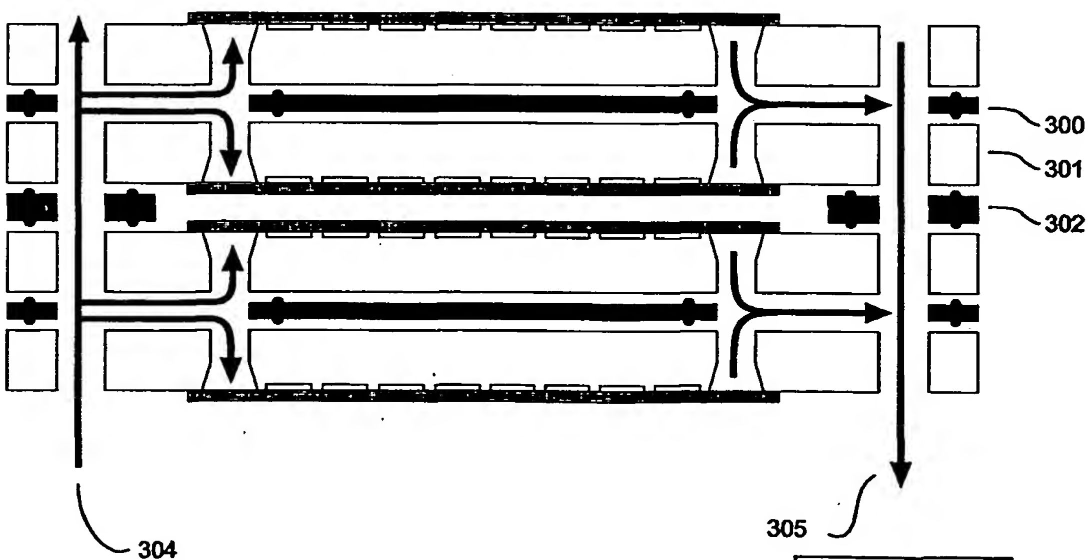

Fig. 27A

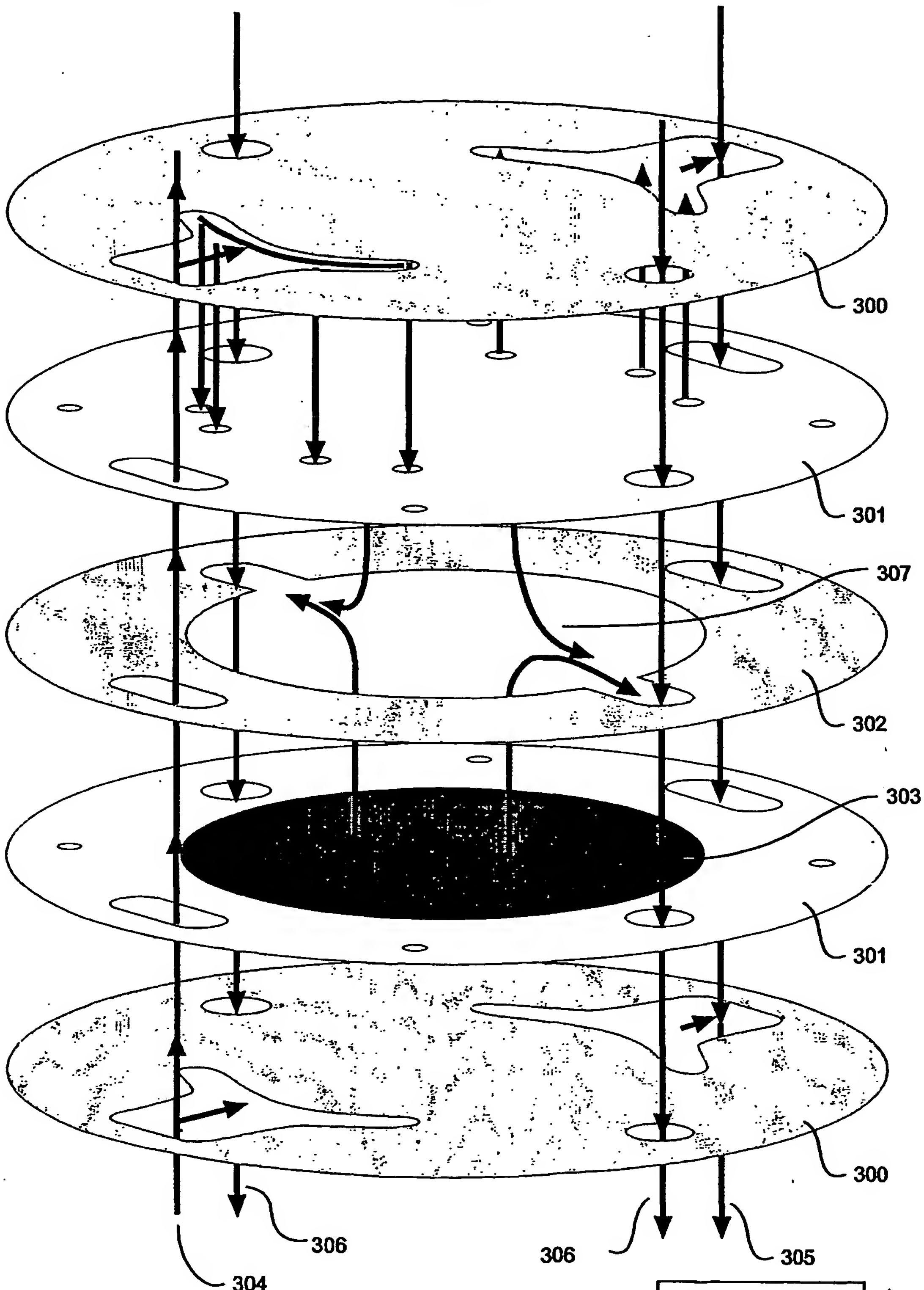

Fig. 27B

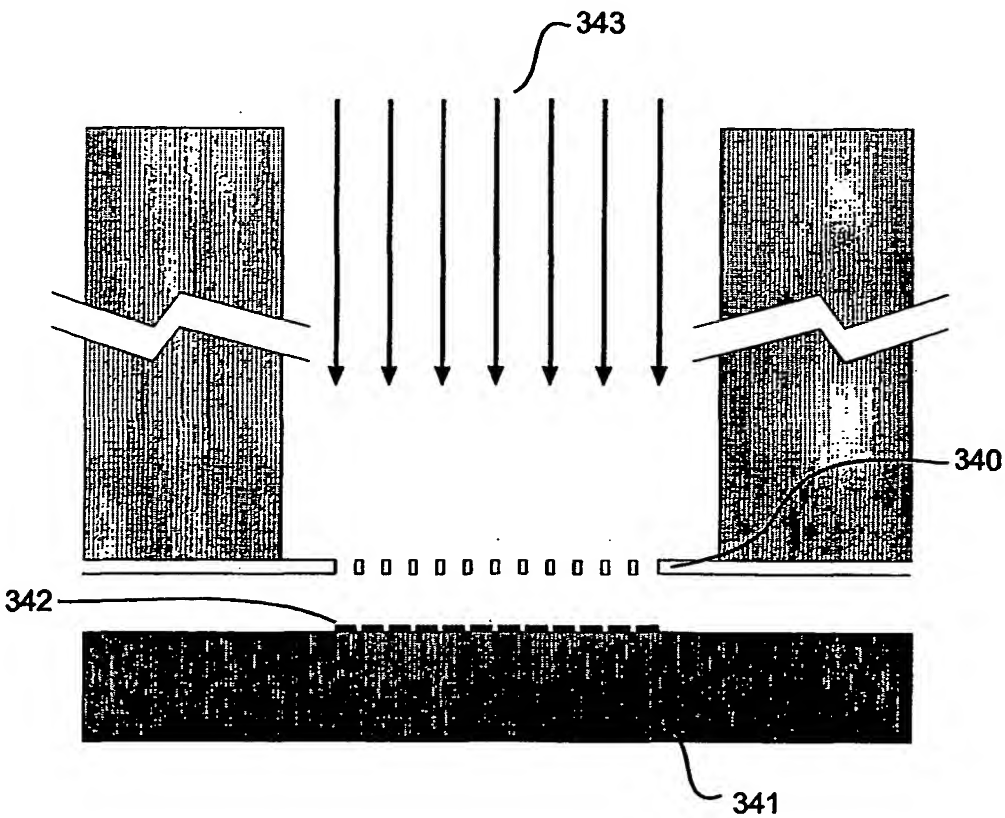

Fig. 28

INTERNATIONAL SEARCH REPORT

Application No
PCT/NL 01/00630

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B05B1/02

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 B05B F02M B41J

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 98 01228 A (SAYLOR JOHN R ;ROVELSTAD AMY L (US); SKEATH PERRY R (US); SPRAYCHI) 15 January 1998 (1998-01-15) page 7, line 23 -page 15, line 11; figures 1-3 page 24, line 14 -page 27, line 14; figures 37-42	1-3,6,7, 20,21, 32,34
X	US 6 086 195 A (BOHORQUEZ JAIME H ET AL) 11 July 2000 (2000-07-11) column 5, line 46 -column 7, line 52; figures 4-6	1-3,6,7, 21,32
X	US 6 016 969 A (TILTON CHARLES ET AL) 25 January 2000 (2000-01-25) column 4, line 33 -column 7, line 8; figures 1-6	1-3,6,21

-/--

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
25 January 2002	04/02/2002

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31–70) 340–2040, Tx. 31 651 epo nl, Fax: (+31–70) 340–3016	Daintith, E

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INTERNATIONAL SEARCH REPORT

Inte   nal Application No
PCT/NL 01/00630

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 925 205 A (HEYSE JOERG ET AL) 20 July 1999 (1999-07-20) column 1, line 20 -column 4, line 67; figures	1-3,6,21
X	US 5 204 690 A (LORENZE JR ROBERT V ET AL) 20 April 1993 (1993-04-20) column 4, line 58 -column 6, line 9; figures 1,2	1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

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International Application No
PCT/NL 01/00630

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